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SOUND ABSORPTION MEASUREMENTS IN GASES

BY

HELENE NARAYANA

A THESIS

SUBMITTED TO THE FACULTY OF GRADUATE STUDIES IN
PARTIAL FULFILMENT OF THE REQUIREMENTS FOR THE
DEGREE OF DOCTOR OF PHILOSOPHY

DEPARTMENT OF PHYSICS

EDMONTON, ALBERTA

JULY, 1966

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BY
J. H. VAN DER KAM

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The undersigned certify that they have read, and recommend to the Faculty of Graduate Studies for acceptance, a thesis entitled "SOUND ABSORPTION MEASUREMENTS IN GASES", submitted by Hélène Narayana in partial fulfilment of the requirements for the degree of Doctor of Philosophy.

ABSTRACT

Apparatus was built for the purpose of measuring the absorption of sound by the pulse technique in gases at low temperatures. The fixed frequency of operation was 3 Mc/sec, and by varying the pressure of gas, absorption measurements were made at a frequency-to-pressure ratio ranging from 3 to 45 Mc/sec-atm. No reports appear in the literature about the use of the pulse technique at low temperatures.

The instrument was tested for proper operation by measuring the absorption of sound in helium both at room temperature and at liquid nitrogen temperature.

The absorption of ultrasonic waves was measured, at liquid nitrogen temperature, in normal hydrogen and in a para-enriched mixture of hydrogen.

The theory of sound absorption in fluids is summarized: the first part of the treatment deals with the classical absorption due to viscosity and thermal conductivity, while the second part deals with relaxational absorption due to a slowness of energy exchange between the translational and the internal degrees of freedom of a gas molecule. The review of the theory given here is more general than is customarily given on work in gases.

The results are discussed and compared with similar results obtained by different techniques at different

temperatures.

The relaxation time for the $0 \longrightarrow 2$ transition in hydrogen at 76.7°K is found to be approximately 2.1×10^{-8} sec. Measurements both in normal hydrogen and in para-enriched hydrogen yield the same value of the relaxation time; this is in contrast to the results of Sluijter, Knaap and Beenakker. The theories for calculation of relaxation times are not precise enough to shed light on this discrepancy.

ACKNOWLEDGEMENTS

The author wishes to thank Prof. S. B. Woods who supervised this project. His constant encouragement and hopeful attitude were most appreciated.

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The author is grateful to all members of the technical staff, and in particular to Mr. Hank Mc Clung, for their invaluable help.

Financial assistance from the University of Alberta, as well as from the National Research Council, is gratefully acknowledged.

And finally, to a patient, sympathetic and understanding husband, simply but wholeheartedly: Thank you.

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CHAPTER I

THEORY

Introduction

The propagation of sound in fluids has been a subject of interest for well over a century. The first theoretical treatment of importance on the subject, dates back to 1845. It is due to Stokes, who investigated the effects of viscosity on the propagation of sound. Later, in 1868, Kirchhoff showed that heat conduction in fluids gives rise to an absorption of sound which is of the same order of magnitude as that due to viscosity. A few authors, among whom Lord Rayleigh is prominent, have also evaluated the effect of heat radiation on the propagation of sound in fluids; their estimates showed however, that radiation plays little role in gases, except perhaps, at very low pressures and very high temperatures.

Early experimental works consistently yielded results which indicated that the absorption of sound in gases other than monatomic gases, was much in excess of that predicted by the so-called classical theory.

In 1928, Herzfeld and Rice suggested that the excess absorption of sound in fluids might be due to a phenomenon that they termed "thermal relaxation." Much refined in the following years, this theory has gained wide acceptance, and accounts for a great number of experimental results. Sound absorption data in fluids can yield very valuable information about the nature of the interaction between colliding particles, since the energy transfer depends strongly upon the non-spherical part of that interaction.

A recent treatment of the subject by Bauer,¹ based on the principles of the thermodynamics of irreversible processes, is of particular interest in view of its relatively simple, and surely very elegant solution of such complicated problems as multiple relaxation processes in a three-level gas, or in mixtures of two gases with excitable states.

Therefore, a unified treatment, using the methods of the thermodynamics of irreversible processes, as developed by Meixner, will yield the "classical" sound absorption equations, also known as the Stokes-Navier equations, as well as those for the "anomalous" sound absorption, due to relaxation. Meixner proved that, to a first approximation, the contributions to the absorption of sound from the transport phenomena and those from the internal transitions, are additive. The two problems can thus be treated separately.

Before dealing especially with the topic of sound absorption in gases, a summary of the fundamentals of the method used is given.

1. Fundamentals of the Phenomenological Thermodynamics of Irreversible Processes

The usual inequality of ordinary thermodynamics relates the increase in entropy dS of a system, to its absolute temperature T , and to the heat supplied to it, dQ , namely

$$dS \geq \frac{dQ}{T} \quad . \quad (1-1)$$

(1-1) can be made an equality by writing as follows:

$$dS = \frac{dQ}{T} + d_i S = d_e S + d_i S, \quad (1-2)$$

in which case the differential $d_e S$ represents the entropy supplied to the system by its surroundings, and $d_i S$ represents the entropy produced inside the system; $d_i S$ is always positive.

One of the most important objectives of the thermodynamics of irreversible phenomena is to relate $d_i S$ to the various irreversible phenomena which may occur within the system. In order to do so, it is convenient to use the entropy

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production per unit time and unit volume - often called the entropy source strength - as a unit of irreversibility:

$$\sigma = \frac{d_i S}{dt} \quad (1-3)$$

Combined with (1-2), the equation expressing the first law of thermodynamics,

$$dU = dQ + dW, \quad (1-4)$$

becomes

$$dW = dU - T dS + T d_i S = dW_{rev} + T d_i S \quad (1-5)$$

where dU is the increase in internal energy of the system, and $(dW - dW_{rev})$ is the work expended or the energy dissipated. The general form of the rate of entropy production is

$$\sigma = \sum_i X_i J_i \quad (1-6)$$

where the J_i 's are the "fluxes" denoting the speed with which the irreversible processes occur, and the X_i 's are the thermodynamic "forces" which are related to the non-uniformity of the system (the gradient of the temperature for example).

Let L be a linear transformation from V to V . Then L is called a linear operator on V . If L is a linear operator on V , then L^2 is also a linear operator on V .

$$L^2 = L \circ L$$

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Since, at equilibrium both the fluxes and the forces disappear, the fluxes are surely functions of the forces. For small departures from equilibrium, as a first approximation

$$J_i = \sum_k L_{ik} X_k, \quad (1-7)$$

where

$$L_{ik} = \frac{\partial J_i}{\partial X_k} \quad . \quad (1-8)$$

Therefore

$$\sigma = \sum_{i,k} L_{ik} X_i X_k \quad . \quad (1-9)$$

The coefficients L_{ik} are called the phenomenological coefficients. Since (1-9) is always positive by virtue of the second law of thermodynamics, the matrix $[L_{ik}]$ is positive definite and is symmetric, i.e., $L_{ik} = L_{ki}$.

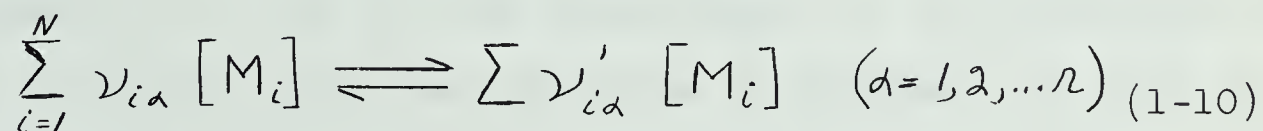
The standard procedure to find the phenomenological coefficients, i.e., to set up equation (1-8), is summarised as follows:

From the rate of change of the entropy, which is determined by the variations in the properties of the system, the entropy source strength σ can be calculated. From (1-9) it is readily noted that σ is a sum of scalar products of the form

$$\frac{\partial J_i}{\partial X_k} X_i X_k.$$

In order to determine the rate of change of the entropy of a system, the state of the system must be defined in terms of certain state variables. These state variables are of two kinds: 1) external (e.g. pressure, temperature) variables which need no further comment and, 2) "chemical" variables if reactions occur which change the material composition of the system. Here the term "chemical" is being used rather loosely; excitations of internal degrees of freedom constitute, in this sense, chemical reactions. Thus the chemical variables need to be examined more carefully. When chemical reactions occur in a system, they can be described in terms of two sets of thermodynamically conjugate variables, called the progress variables and the affinities.

The progress variables are best understood as follows: Suppose there are r possible reactions in a system containing N substances. The reaction equations are given by



where M_i is the mass of substance i , $\nu_{i\alpha}$ and $\nu'_{i\alpha}$ are the left and right stoichiometric coefficients of substance i in the reaction α . When the reaction α proceeds by a differential amount, dn_i^α the change in the number of moles of substance i is given by

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$$\lim_{n \rightarrow \infty} \left(\frac{1}{n} \sum_{k=1}^n \frac{1}{k} \right) = \frac{1}{2}$$

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$$\frac{dn_i^\alpha}{\Delta \nu_{i\alpha}} = \frac{dn_2^\alpha}{\Delta \nu_{2\alpha}} = \text{-----} = d\xi_\alpha \quad (1-11)$$

where $\Delta \nu_{i\alpha} = \nu'_{i\alpha} - \nu_{i\alpha}$. The total change dn_i becomes

$$dn_i = \sum_{\alpha=1}^n \Delta \nu_{i\alpha} d\xi_\alpha \quad (1-12)$$

$d\xi_\alpha$ represents the differential amount by which reaction α has advanced. The parameter ξ_α is known as the progress variable. When the reaction has proceeded to completion to the right, $d\xi_\alpha = 1$, i.e. $\Delta \nu_{i\alpha}$ molecules of substance i have been produced.

As for the affinities, well before a practical measure of these somewhat "anthropomorphic quantities" was found (Joos), the concept of chemical affinity had been widely used. Van't Hoff first recognised that for reactions at constant V and T , the free energy F is the proper measure of affinity, while for reactions at constant p and T , the Gibb's potential G serves this purpose. These quantities are minimum at equilibrium; the greater the values of these functions compared with the minimum values, the further the system is from equilibrium, and consequently, the more readily the reaction will occur.

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If only expansion work is performed on a system, all reactions then being fixed, the process is reversible. The quantity $-pdV$ is then the contribution of the external variables to dW_{rev} . If however, reactions occur at constant volume, the process is not reversible. Therefore other terms must be subtracted from $-pdV$ in order to obtain dW_{rev} . These terms will be of the form

$$\sum_{\alpha} A_{\alpha} d\xi_{\alpha} ,$$

where the A_{α} 's are the affinities, and the ξ_{α} 's are the progress variables. Therefore

$$dW_{\text{rev}} = -pdV - \sum_{\alpha} A_{\alpha} d\xi_{\alpha} .$$

The first law of thermodynamics thus takes the form

$$dU = dW_{\text{rev}} + dQ_{\text{rev}} = T dS - pdV - \sum_{\alpha} A_{\alpha} d\xi_{\alpha}, \quad (1-13)$$

from which we obtain an expression for A_{α} :

$$A_{\alpha} = -\left(\frac{\partial U}{\partial \xi_{\alpha}}\right)_{VS} \quad (1-14)$$

Just as $-p$ and T are thermodynamically conjugate to V and S respectively, $-A$ is thermodynamically conjugate to ξ_α .

Since the various thermodynamic potentials are defined by the equations

$$G = U - TS + pV$$

$$F = U - TS$$

and $H = U + pV,$

the following expressions for A also hold:

$$A_\alpha = -\left(\frac{\partial G}{\partial \xi_\alpha}\right)_{pT} = -\left(\frac{\partial F}{\partial \xi_\alpha}\right)_{VT} = -\left(\frac{\partial H}{\partial \xi_\alpha}\right)_{pS} \quad (1-15)$$

where G , F , H are respectively the Gibbs free enthalpy, the Helmholtz free energy and the enthalpy.

The physical variables x and y will now be introduced. The quantity y stands for either S or T , while x stands for either V or $-p$. The conjugate variables will be expressed by X and Y . Finally P denotes the thermodynamic potential corresponding to x and y .

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$$\int_0^1 (x^2 + 1) dx = \left[\frac{x^3}{3} + x \right]_0^1 = \frac{1}{3} + 1 = \frac{4}{3}$$

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Equation (1-13) takes the form: ($x=V$, $X=p$,
 $y=S$, $Y=T$)

$$dP = -X dx + Y dy - \sum_{\alpha} A_{\alpha} d\xi_{\alpha} \quad (1-16)$$

while equation (1-15) becomes

$$A_{\alpha} = - \left(\frac{\partial P}{\partial \xi_{\alpha}} \right)_{xy} \quad (1-17)$$

Differentiating A_{α} with respect to x yields:

$$\left(\frac{\partial A_{\alpha}}{\partial x} \right)_{\xi y} = - \left(\frac{\partial^2 P}{\partial \xi_{\alpha} \partial x} \right)_y = \left(\frac{\partial X}{\partial \xi_{\alpha}} \right)_{xy} \quad (1-18)$$

A few other useful formulae can be derived, and are given below.

$$\left(\frac{\partial A_{\alpha}}{\partial x} \right)_{\xi y} = - \left(\frac{\partial A_{\alpha}}{\partial \xi_{\alpha}} \right)_{xy} \left(\frac{\partial \xi_{\alpha}}{\partial x} \right)_{A_{\alpha} y} \quad (1-19a)$$

$$\left(\frac{\partial A_{\alpha}}{\partial \xi_{\alpha}} \right)_{xy} = \left(\frac{\partial A_{\alpha}}{\partial \xi_{\alpha}} \right)_{xy} \left(\frac{\partial x}{\partial X} \right)_{\xi y} \left(\frac{\partial X}{\partial x} \right)_{A_{\alpha} y} \quad (1-19b)$$

The fundamentals of the thermodynamics of irreversible processes have been summarized in this section, following the treatment by Bauer (1965).

2. Classical Sound Absorption

Using the principles stated in the preceding section, the rate of entropy production will be determined, showing that, in the case of an isotropic fluid without reactions, the fluxes are the heat flux \vec{J}_q and the pressure tensor Π , while their respective conjugated forces are the gradient of the reciprocal temperature, and the flow velocities. The phenomenological equations will then be set up, which together with the equations of continuity for mass, momentum and energy, constitute a system of homogeneous equations. When this system of equations is linearized and solved, the complex wave vector k for the sound propagation can be found. A detailed exposition for finding the wave vector k now follows.

By the principle of conservation of energy, the total energy content within an arbitrary volume V in the system can only change if energy flows into or out of the volume through its surface. With the help of Gauss' theorem, the differential form of the law of conservation of energy is obtained:

$$\frac{\partial \rho e}{\partial t} = - \nabla \cdot \vec{J}_e , \quad (1-25)$$

where ρ is the density, e is the total energy per unit mass and \vec{J}_e the total energy flux per unit surface and unit time. Here e includes all forms of energy in the system and may be written in the form,

$$e = \frac{1}{2} v^2 + u, \quad (1-26)$$

where $\frac{1}{2} v^2$ is the specific kinetic energy, and u is the specific internal energy. Similarly \vec{J}_e includes all forms of energy fluxes: a convective term $\rho e \vec{v}$, an energy flux $\vec{\Pi} \cdot \vec{v}$ due to the mechanical work performed on the system, and finally, the heat flux \vec{J}_q :

$$\vec{J}_e = \rho e \vec{v} + \vec{\Pi} \cdot \vec{v} + \vec{J}_q . \quad (1-27)$$

The conservation law for energy of a fluid in the absence of external forces can thus be written:

$$\rho \frac{de}{dt} = - \nabla \cdot (\vec{\Pi} \cdot \vec{v} + \vec{J}_q) \quad (1-28)$$

where

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \vec{v} \cdot \nabla \quad (1-29)$$

is the so-called differential following the motion.

Remembering that in a fluid, only differences of pressure give rise to a force (constant, normal or shearing stresses do not produce a force) the conservation law of momentum can be written as

THEORY

The first part of the theory is the definition of the function $f(x)$. This function is defined as the sum of the squares of the first n natural numbers. The second part of the theory is the proof that the function $f(x)$ is a polynomial of degree 3. The third part of the theory is the proof that the function $f(x)$ is a polynomial of degree 3.

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$$\rho \frac{d\vec{v}}{dt} = - \nabla \cdot \Pi \quad . \quad (1-30)$$

Combining equations (1-26) and (1-28) gives the balance of internal energy.

$$\rho \frac{du}{dt} = - \Pi : \nabla \vec{v} - \nabla \cdot \vec{J}_q \quad (1-31)$$

It is now useful to split the pressure tensor into two parts: the hydrostatic pressure p , and the symmetric viscous pressure tensor Π_s . This is justified, since the components of the stress tensor are the following:

$$\Pi_{ii} = p - 2\eta \left(\frac{\partial v_i}{\partial r_i} - \frac{1}{3} \nabla \cdot \vec{v} \right) \quad (1-32a)$$

$$\Pi_{ij} = \eta \left(\frac{\partial v_i}{\partial r_j} + \frac{\partial v_j}{\partial r_i} \right) \quad (1-32b)$$

where $r_i, r_j = x, y, z$

$v_i, v_j = v_x, v_y, v_z$

and η is the shear viscosity. For a monatomic gas, the bulk viscosity is identically equal to zero. The hydrostatic pressure

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[illegible]

He is now residing at [illegible]

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25-12-1918 31-12-1918

The above named person was born on 10th March 1880 at [illegible]
[illegible]

[illegible]

$$p = \frac{1}{3} (\pi_{xx} + \pi_{yy} + \pi_{zz}). \quad (1-33)$$

The balance equation for the internal energy and the law of conservation of mass become respectively:

$$\rho \frac{du}{dt} = -p \nabla \cdot \vec{v} - \pi_s : \nabla \vec{v} - \nabla \cdot \vec{J}_q, \quad (1-34)$$

and

$$\rho \frac{dV}{dt} = \nabla \cdot \vec{v}. \quad (V \equiv \rho^{-1}) \quad (1-35)$$

From the last two equations,

$$\rho \left(\frac{du}{dt} + p \frac{dV}{dt} \right) = -\pi_s : \nabla \vec{v} - \nabla \cdot \vec{J}_q \quad (1-36)$$

which can be inserted into the Gibbs relation:

$$T \frac{dS}{dt} = \frac{du}{dt} + p \frac{dV}{dt} \quad (1-37)$$

Thus the entropy balance equation is obtained:

$$\rho \frac{dS}{dt} = -\nabla \cdot \vec{J}_s + \sigma \quad (1-38)$$

with the entropy flow

$$\vec{J}_s = \frac{\vec{J}_q}{T} \quad (1-39)$$

Let $\mathcal{A} = \{A_1, A_2, \dots, A_n\}$ be a family of subsets of S .

Suppose that for every $s \in S$, the number of sets in \mathcal{A} containing s is at most k .

Prove that $|\mathcal{A}| \leq \frac{|S|}{k}$.

Hint: Consider the sum $\sum_{s \in S} |\{A \in \mathcal{A} : s \in A\}|$.

Use the inequality $\sum_{i=1}^n a_i \leq n \cdot \max\{a_1, a_2, \dots, a_n\}$.

Let $f: \mathbb{R} \rightarrow \mathbb{R}$ be a function satisfying

$$f(x+y) = f(x) + f(y) \quad \text{for all } x, y \in \mathbb{R}.$$

Suppose also that f is continuous at 0 .

Prove that $f(x) = cx$ for some constant $c \in \mathbb{R}$.

Hint: First show that $f(x) = cx$ for all rational x .

Then use the continuity of f at 0 .

Let \mathcal{A} be a family of subsets of S such that

and the entropy production

$$\sigma = - \vec{J}_q \cdot \frac{\nabla(T)}{T^2} - \frac{\pi_s : \nabla \vec{v}}{T} \geq 0 \quad (1-40)$$

As mentioned before, the entropy production is a sum of products of fluxes and thermodynamic forces, the former being related to the latter through the phenomenological equations (1-7). Since the phenomenological coefficients can be temperature dependent, new coefficients can now be introduced into which such factors as $\frac{1}{T}$ and $\frac{1}{T^2}$ can be absorbed.

The familiar expression

$$\vec{J}_q = - \varphi \nabla T, \quad (1-41)$$

where φ is the heat conductivity, is obtained through such simple considerations, from equation (1-40). Similarly, equations (1-32) yield in turn

$$\pi_s = -2\eta \left(\nabla \vec{v} - \frac{1}{3} \nabla \cdot \vec{v} U \right), \quad (1-42)$$

where U is a unit tensor.

The present treatment follows in broad outline and is a simplification of the treatment by De Groot and Mazur², but will henceforth be restricted to one dimension. In such a case, the laws of conservation of mass and momentum are

$$\frac{d\rho}{dt} + \rho \frac{\partial v_x}{\partial x} = 0 \quad (1-43)$$

and

$$\rho \frac{dv_x}{dt} + \frac{\partial p}{\partial x} - \frac{4}{3} \eta \frac{\partial^2 v_x}{\partial x^2} = 0 \quad (1-44)$$

Again, restricting the problem to one-dimension, and inserting the phenomenological equations (1-41), (1-42) into the energy law (1-34), the following expression is obtained:

$$\rho \frac{du}{dt} + p \frac{\partial v_x}{\partial x} - \frac{4}{3} \eta \left(\frac{\partial v_x}{\partial x} \right)^2 - \kappa \frac{\partial^2 T}{\partial x^2} = 0 \quad (1-45)$$

Now it is well known that for a monatomic gas whose molecules possess translational kinetic energy only, the internal energy of the gas equals $\frac{3}{2} kT$. Moreover, from the equation of state of the gas (k is Boltzmann's constant)

$$T = \frac{p}{\rho} \left(\frac{m}{k} \right) \quad (1-46)$$

The following is a list of the names of the persons who have been elected to the office of the President of the United States, and the names of the persons who have been elected to the office of the Vice President of the United States, in the year 1800.

John Adams, President of the United States.

Thomas Jefferson, Vice President of the United States.

The following is a list of the names of the persons who have been elected to the office of the President of the United States, and the names of the persons who have been elected to the office of the Vice President of the United States, in the year 1804.

James Madison, President of the United States.

The following is a list of the names of the persons who have been elected to the office of the President of the United States, and the names of the persons who have been elected to the office of the Vice President of the United States, in the year 1808.

James Madison, President of the United States.

where m is the mass of a molecule of the gas. Therefore the internal energy per unit mass becomes

$$u = \frac{3}{2} \frac{p}{\rho} \quad . \quad (1-47)$$

The three preceding equations have to be linearized, in order to handle them easily mathematically. In the case of moderately weak ultrasonic waves, the usual procedure for linearizing these equations is to express ρ and p as

$$\rho = \rho_0 (1 + \epsilon) \quad , \quad p = p_0 (1 + \delta) \quad . \quad (1-48)$$

Here ρ_0 and p_0 are respectively the density and the pressure of the fluid in the absence of perturbation. ϵ and δ are small compared to unity, while \vec{v} the flow velocity is very much smaller than the sound velocity. Therefore, powers higher than one and products of these quantities or of their derivatives, can be neglected. Substituting for ρ , p and u , equations (1-43), (1-44) and (1-45) become:

$$\frac{\partial \epsilon}{\partial t} + \frac{\partial v_x}{\partial x} = 0 \quad (1-49)$$

$$\frac{\partial v_x}{\partial t} + \frac{p_0}{\rho_0} \frac{\partial \delta}{\partial x} - \frac{4}{3} \frac{\eta}{\rho_0} \frac{\partial^2 v_x}{\partial x^2} = 0 \quad \text{and} \quad (1-50)$$

$$\frac{3}{2} \frac{\partial}{\partial t} (\delta - \epsilon) + \frac{\partial v_x}{\partial x} - \frac{\eta}{\rho_0 k} \frac{\partial^2}{\partial x^2} (\delta - \epsilon) = 0 \quad . \quad (1-51)$$

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REPORT OF THE DEPARTMENT OF CHEMISTRY
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Eliminating v_x from the above equations, there results

$$\frac{p_r}{\rho_0} \frac{\partial^2 \delta}{\partial x^2} - \frac{\partial^2 \epsilon}{\partial t^2} + \frac{4}{3} \frac{\eta}{\rho_0} \frac{\partial^3 \epsilon}{\partial x^2 \partial t} = 0 \quad (1-52)$$

and

$$\frac{\partial \delta}{\partial t} - \frac{5}{3} \frac{\partial \epsilon}{\partial t} - \frac{2}{3} \left(\frac{\varphi}{\rho_0 k} \right) \frac{\partial^2}{\partial x^2} (\delta - \epsilon) = 0 \quad (1-53)$$

Equations (1-52) and (1-53) are the equations governing the propagation of sound waves of small amplitude in a gas treated as a continuous medium. Assuming that the sound wave is propagated as a damped progressive wave, ϵ and δ must be of the form

$$\epsilon = \epsilon_0 e^{i(Kx - \omega t)} \quad \text{and} \quad \delta = \delta_0 e^{i(Kx - \omega t)} .$$

Substituting these values into equations (1-52), (1-53) yields two homogeneous equations for ϵ_0 and δ_0 .

$$\epsilon_0 \left(\omega^2 + i \frac{4}{3} \frac{\eta}{\rho_0} \omega K^2 \right) - K^2 \frac{p_r}{\rho_0} \delta_0 = 0 \quad (1-54)$$

$$\epsilon_0 \left(\frac{5}{3} + i \frac{2}{3} \frac{\varphi}{\rho_0} \frac{K^2}{k \omega} \right) + \delta_0 \left(-i \frac{2}{3} \frac{\varphi}{\rho_0} \frac{K^2}{k} - 1 \right) = 0 \quad (1-55)$$

Since the determinant of these equations must be zero for consistency, we obtain a quadratic equation in K^2 , namely

$$K^4 \left[-\frac{8}{9} \frac{\eta}{\rho_0^2} \frac{\omega}{k} + i \frac{2}{3} \frac{\varphi}{\rho_0} \frac{p_0}{k\omega} \right] + K^2 \left[\frac{5}{3} \frac{p_0}{\rho_0} + i \frac{\omega^2 \varphi}{3 \rho_0 k} \right. \\ \left. + i \frac{4}{3} \frac{\eta}{\rho_0} \omega \right] + \omega^2 = 0 \quad (1-56)$$

An approximation to the classical sound absorption coefficient α_{class} , which is often encountered in the literature can be derived from equation (1-56) by means of various substitutions. Firstly

$$v_0 = \left(\frac{5}{3} \frac{p_0}{\rho_0} \right)^{1/2}$$

is the adiabatic sound velocity at low frequencies for a monatomic gas. Next let $k_i = \frac{K}{\beta_0} = \frac{K v_0}{\omega}$ and

$$\varphi = \frac{15}{4} k \quad . \quad \text{Further, following Greenspan}^3$$

let $R = \frac{\rho_0 v_0^2}{\eta \omega}$ where R is the Reynold's number taken from the field of gas dynamics. Equation (1-56) becomes

$$k_i^4 \left[\frac{3}{2} \frac{i}{R} - \frac{10}{3} \frac{1}{R^2} \right] + k_i^2 \left[1 + \frac{23}{6} \frac{i}{R} \right] + 1 = 0$$

$$\text{or} \quad k_i^2 = R \frac{R + \frac{23}{6} i - \left(R^2 - \frac{49}{36} + \frac{5}{3} i R \right)^{1/2}}{\frac{20}{3} - 3 i R}$$

$$\text{Thus} \quad k_i = i + \frac{7}{6} \frac{1}{R} - \frac{141}{72} \frac{i}{R^2} - \frac{1559}{432} \frac{1}{R^3} + \dots \quad (1-57)$$

which is obtained from the preceding equation by series approximation.

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In the zeroth approximation, the velocity is constant, and there is no attenuation. The first approximation gives the "classical" attenuation per unit length

$$\frac{\alpha_{\text{class}}}{\beta_0} = \frac{\alpha_{\text{class}} \lambda_0}{2\pi} = \frac{7}{6} \frac{1}{R}, \quad (1-58)$$

where λ_0 is the wave-length at low frequencies. Equation (1-58) can be shown to be equivalent to the usual approximation:

$$\alpha_{\text{class}} = \frac{2\pi^2}{\gamma p_0 V_0} \left[\frac{4}{3} \eta + \frac{\gamma-1}{C_p} \phi \right] b^2 \quad (1-59)$$

Here C_p is the specific heat at constant pressure, γ is the ratio of the specific heats and f is the number of vibrations per unit time of the sound wave.

Indeed the equivalence of (1-58), (1-59) follows easily, since $\gamma = \frac{5}{3}$ in the case of a monatomic gas and $\lambda = \frac{5}{2} \eta C_v$ by the kinetic theory of gases, where C_v is the specific heat at constant volume. Thus the classical sound absorption is seen from (1-59), to be directly proportional to the square of the frequency, and inversely proportional to the pressure.

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3. Relaxational Absorption

When a fluid is given a periodic deformation, as is the case when a sound wave is transmitted through the fluid, a phase lag exists between the stresses and the strains, which is responsible for the absorption of sound. In monatomic gases, this phase lag is due entirely to viscosity and heat conduction. In diatomic and polyatomic gases however, the absorption of sound is also due to the failure of the internal degrees of freedom to remain in thermal equilibrium with the external degrees of freedom. The process by which the temperature of the internal degrees of freedom adjusts itself to the temperature of the external degrees of freedom is a typical relaxation process. Due to the passage of the sound wave, the "translational temperature" is increased. In order to be again in equilibrium with the new translational energy, the internal energy must also increase; i.e. energy must flow from the translational to the internal degrees of freedom. This can only occur through collisions, and takes a finite time. The time needed to decrease the maladjustment by a factor e^{-1} is called the relaxation time. Such a process is called a reaction in the language of the thermodynamics of irreversible processes and the value of the relaxation time depends on whether the reaction proceeds isothermally or adiabatically.

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Again, following Bauer (1965), the phenomenological equations are easily set up as follows. Combining equations (1-3), (1-5) and (1-13) yields the rate of entropy production,

$$\sigma = \frac{1}{T} \sum A_{\alpha} \dot{\xi}_{\alpha} .$$

After absorbing the factor $\frac{1}{T}$ into the phenomenological coefficients, the phenomenological equations become:

$$\dot{\xi}_{\alpha} = \sum_{\beta} L_{\alpha\beta} A_{\beta} \quad (\alpha = 1, \dots, r). \quad (1-60)$$

In the case of a single relaxation process, equation (1-60) takes the form:

$$\dot{\xi} = LA \quad . \quad (1-61)$$

L may be assumed to be constant within a small region. However A depends on the variables $x(\equiv V$ or $-p)$, $y(\equiv S$ or $T)$ and ξ . Within a small region about the point (x_0, y_0, ξ_0) :

$$\begin{aligned} A(x, y, \xi) = & A(x_0, y_0, \xi_0) + \left(\frac{\partial A}{\partial \xi} \right)_{xy} (\xi - \xi_0) \\ & + \left(\frac{\partial A}{\partial x} \right)_{\xi y} (x - x_0) + \left(\frac{\partial A}{\partial y} \right)_{\xi x} (y - y_0) . \end{aligned} \quad (1-62)$$

If the point (x_0, y_0, ξ_0) is an equilibrium point, $A(x_0, y_0, \xi_0)$ vanishes, since as stated earlier, the affinities vanish at equilibrium. From equations (1-60), (1-61) the so-called relaxation equation (1-62) is obtained.

$$\dot{\xi} = L \left[\left(\frac{\partial A}{\partial \xi} \right)_{xy} (\xi - \xi_0) + \left(\frac{\partial A}{\partial \mu} \right)_{\xi y} (\mu - \mu_0) + \left(\frac{\partial A}{\partial y} \right)_{\xi \mu} (y - y_0) \right] \quad (1-63)$$

Moreover by holding the external variables x and y constant at $x = x_0$, $y = y_0$ the last equation becomes a linear homogeneous differential equation whose solution is

$$(\xi - \xi_0) = (\xi_{t=0} - \xi_0) \exp \left(-\frac{t}{\tau_{xy}} \right) \quad (1-64)$$

with the reciprocal relaxation time

$$\frac{1}{\tau_{xy}} = - L \left(\frac{\partial A}{\partial \xi} \right)_{xy} . \quad (1-65)$$

If the experiment is performed under different conditions, so that X and y are kept constant, rather than x and y , a different relaxation time τ_{xy} will result. The relation between these various relaxation times is given by equations (1-65) and (1-19b)

$$\frac{\hat{\tau}_{xy}}{\hat{\tau}_{xy}} = \left(\frac{\partial A}{\partial \xi} \right)_{xy} \bigg/ \left(\frac{\partial A}{\partial \xi} \right)_{xy} = \left(\frac{\partial \kappa}{\partial X} \right)_{\xi y} \left(\frac{\partial X}{\partial \kappa} \right)_{Ay}$$

the operator $\left(\frac{\partial}{\partial \kappa} \right)_{Ay}$ represents the differentiation carried out during equilibrium, while the operator $\left(\frac{\partial}{\partial \kappa} \right)_{\xi y}$ represents the differentiation carried out during an arrested reaction. The values obtained by means of these operators are often referred to as quasi-static and instantaneous or frozen-in values respectively.

For the four variables p , V , T and S , the various relaxation times are related as follows:

$$\frac{\hat{\tau}_{pT}}{\hat{\tau}_{VT}} = \left(\frac{\partial A}{\partial \xi} \right)_{VT} \bigg/ \left(\frac{\partial A}{\partial \xi} \right)_{pT} = \left(\frac{\partial V}{\partial p} \right)_{AT} \left(\frac{\partial p}{\partial V} \right)_{\xi T} = K_T^o / K_T^\infty, \quad (1-66)$$

$$\frac{\hat{\tau}_{pT}}{\hat{\tau}_{pS}} = \left(\frac{\partial A}{\partial \xi} \right)_{pS} \bigg/ \left(\frac{\partial A}{\partial \xi} \right)_{pT} = \left(\frac{\partial S}{\partial T} \right)_{Ap} \left(\frac{\partial T}{\partial S} \right)_{\xi p} = C_p^o / C_p^\infty, \quad (1-67)$$

$$\frac{\hat{\tau}_{VT}}{\hat{\tau}_{VS}} = \left(\frac{\partial A}{\partial \xi} \right)_{VS} \bigg/ \left(\frac{\partial A}{\partial \xi} \right)_{VT} = \left(\frac{\partial S}{\partial T} \right)_{AV} \left(\frac{\partial T}{\partial S} \right)_{\xi V} = C_v^o / C_v^\infty, \quad (1-68)$$

$$\frac{\hat{\tau}_{pS}}{\hat{\tau}_{VS}} = \left(\frac{\partial A}{\partial \xi} \right)_{VS} \bigg/ \left(\frac{\partial A}{\partial \xi} \right)_{pS} = \left(\frac{\partial V}{\partial p} \right)_{AS} \left(\frac{\partial p}{\partial V} \right)_{\xi S} = K_s^o / K_s^\infty. \quad (1-69)$$

K_T and K_S are the isothermal and the adiabatic compressibilities respectively. The indices o and ∞ refer to the quasi-static and instantaneous values respectively.

Unfortunately, the progress variables cannot be observed themselves, and the relaxation times cannot be determined as easily as equation (1-64) seems to indicate. They can only be found from the behaviour of another dependent variable z , which is a function of x , y and ξ . Therefore, if instead of being kept constant, x and y are allowed to vary, with the help of (1-65) and (1-19a), equation (1-63) becomes:

$$\dot{\xi} = -\frac{1}{\tau_{xy}} \left[(\xi - \xi_0) - \left(\frac{\partial \xi}{\partial x} \right)_{Ay} (x - x_0) - \left(\frac{\partial \xi}{\partial y} \right)_{Ax} (y - y_0) \right] \quad (1-70)$$

On the other hand $z = z(x, y, \xi)$ can be expanded linearly about x_0, y_0, ξ_0 as follows:

$$z - z_0 = \left(\frac{\partial z}{\partial \xi} \right)_{xy} (\xi - \xi_0) + \left(\frac{\partial z}{\partial x} \right)_{\xi y} (x - x_0) + \left(\frac{\partial z}{\partial y} \right)_{\xi x} (y - y_0) \quad (1-71)$$

$\xi, \dot{\xi}$ can be eliminated from the last two equations, yielding

$$\begin{aligned} \left[1 + \tau_{xy} \frac{d}{dt} \right] (z - z_0) &= \left[\left(\frac{\partial z}{\partial \xi} \right)_{xy} \left(\frac{\partial \xi}{\partial x} \right)_{Ay} + \left(\frac{\partial z}{\partial x} \right)_{\xi y} \left(1 + \tau_{xy} \frac{d}{dt} \right) \right] (x - x_0) \\ &+ \left[\left(\frac{\partial z}{\partial \xi} \right)_{xy} \left(\frac{\partial \xi}{\partial y} \right)_{Ax} + \left(\frac{\partial z}{\partial y} \right)_{\xi x} \left(1 + \tau_{xy} \frac{d}{dt} \right) \right] (y - y_0) \end{aligned} \quad (1-72)$$

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From equation (1-19b), it follows that

$$\left(\frac{\partial z}{\partial \xi}\right)_{xy} \left(\frac{\partial \xi}{\partial \kappa}\right)_{Ay} = \left(\frac{\partial z}{\partial \kappa}\right)_{Ay} - \left(\frac{\partial z}{\partial \kappa}\right)_{\xi y} ,$$

and that

$$\left(\frac{\partial z}{\partial \xi}\right)_{xy} \left(\frac{\partial \xi}{\partial y}\right)_{Ax} = \left(\frac{\partial z}{\partial y}\right)_{Ax} - \left(\frac{\partial z}{\partial \kappa}\right)_{\xi y} .$$

Equation (1-72) now becomes:

$$\begin{aligned} \left[1 + \tau_{xy} \frac{d}{dt}\right] (z - z_0) &= \left[\left(\frac{\partial z}{\partial \kappa}\right)_{Ay} + \left(\frac{\partial z}{\partial \kappa}\right)_{\xi y} \tau_{xy} \frac{d}{dt} \right] (\kappa - \kappa_0) \\ &\quad + \left[\left(\frac{\partial z}{\partial y}\right)_{Ax} + \left(\frac{\partial z}{\partial y}\right)_{\xi \kappa} \tau_{xy} \frac{d}{dt} \right] (y - y_0) . \end{aligned} \quad (1-73)$$

Assuming that the state variables are of the form:

$$(x - x_0) = C e^{j(\omega t + d)} ,$$

the differential operator $\frac{d}{dt}$ is simply $j\omega$. Thus equation (1-73) takes on the form

$$\begin{aligned} (z - z_0) &= \left[\left(\frac{\partial z}{\partial \kappa}\right)_{Ay} + j\omega \tau_{xy} \left(\frac{\partial z}{\partial \kappa}\right)_{\xi y} (\kappa - \kappa_0) + \left(\frac{\partial z}{\partial y}\right)_{Ax} \right. \\ &\quad \left. + j\omega \tau_{xy} \left(\frac{\partial z}{\partial y}\right)_{\xi \kappa} (y - y_0) \right] / (1 + j\omega \tau_{xy}) \end{aligned}$$

or

$$(z - z_0) = \left(\frac{\partial z}{\partial x} \right)_y^{eff} (x - x_0) + \left(\frac{\partial z}{\partial y} \right)_x^{eff} (y - y_0) ,$$

where

$$\left(\frac{\partial z}{\partial x} \right)_y^{eff} = \left(\frac{\partial z}{\partial x} \right)_{Ay} \frac{1 + j\omega \hat{\tau}_{yz}}{1 + j\omega \hat{\tau}_{xy}} \quad (1-74a)$$

and

$$\left(\frac{\partial z}{\partial y} \right)_x^{eff} = \left(\frac{\partial z}{\partial y} \right)_{Ax} \frac{1 + j\omega \hat{\tau}_{xz}}{1 + j\omega \hat{\tau}_{xy}} . \quad (1-74b)$$

The derivatives $\left(\frac{\partial z}{\partial x} \right)_y^{eff}$ and $\left(\frac{\partial z}{\partial y} \right)_x^{eff}$ are clearly the effective state coefficients, which take into account the finite reaction rates. These equations can now be applied to the case of acoustic relaxation.

If we neglect the transport phenomena, the adiabatic character of the process is guaranteed. In the absence of the dissipative factors, the equations of continuity for momentum, mass and energy yield the Laplacian formula for the wave-vector k_0 :

$$\frac{k_0^2}{\omega^2} = K_S^0 \rho = \frac{1}{c_0^2} \quad (1-75)$$

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where K_S^0 is the adiabatic compressibility, in the absence of relaxation. If the internal transitions are now taken into account, the adiabatic compressibility must be replaced by an effective adiabatic compressibility K_S^{eff} . K_S^{eff} can be easily found in terms of τ_{yz} and τ_{xy} . To do this the following choice of variables is made:

$$x \equiv -p, \quad y \equiv S \equiv S_0, \quad z = \rho = \frac{M}{V}.$$

Using equation (1-74) with

$$\frac{1}{\rho} \left(\frac{\partial \rho}{\partial p} \right)_S = -\frac{1}{V} \left(\frac{\partial V}{\partial p} \right)_S = K_S \quad \text{and} \quad \tau_{\rho S} \equiv \tau_{VS},$$

a "normalized" effective adiabatic compressibility is obtained:

$$\frac{K_S^{\text{eff}}}{K_S^0} = \frac{1 + j\omega \tau_{VS}}{1 + j\omega \tau_{\rho S}}.$$

The locus of K_S^{eff}/K_S^0 can be obtained by means of the conformal mapping $w = \frac{1 + j\tau_{VS}}{1 + j\tau_{\rho S}}$ onto the positive real axis $z = 1/\omega$. This gives a semi-circle of diameter $\epsilon = 1 - \frac{\tau_{VS}}{\tau_{\rho S}}$.

ϵ is called the relaxation strength. The maximum of the imaginary part occurs when $\omega = \frac{1}{\tau_{\rho S}}$. This locus, which is known as the Cole plot, is useful in determining

whether sound absorption or dispersion is due to a single relaxation process or to a multiple relaxation process: the curve is a semi-circle in the former case only.

Under these conditions, equation (1-75) must be replaced by:

$$\frac{k_r^2}{\omega^2} = K_S^{\text{eff}} \rho \quad (1-76)$$

where k_r is the complex wave vector

$$k_r = \frac{\omega}{C} - j \alpha' \quad (1-77)$$

If the value of k_r given by (1-77) is substituted into (1-76), and both sides are multiplied by $C_o^2 = \frac{1}{\rho K_s}$ an obvious simplification yields

$$\begin{aligned} \frac{C_o^2}{C^2} - \frac{\alpha' C_o^2}{\omega^2} - j \frac{2\alpha' C_o^2}{\omega C} &= \frac{K^{\text{eff}}}{K_s} \\ &= \frac{1 + \omega^2 \hat{\tau}_{vs} \hat{\tau}_{ps}}{1 + \omega^2 \hat{\tau}_{ps}^2} - j \frac{\omega (\hat{\tau}_{ps} - \hat{\tau}_{vs})}{1 + \omega^2 \hat{\tau}_{ps}^2} \end{aligned} \quad (1-78)$$

The real and imaginary parts of (1-78) will be equated separately in what follows. Comparing first the real part,

$$\frac{C_o^2}{C^2} - \frac{\alpha' C_o^2}{\omega^2} = \frac{1 + \omega^2 \hat{\tau}_{ps} \hat{\tau}_{vs}}{1 + \omega^2 \hat{\tau}_{ps}^2} \quad (1-79)$$

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Now, C_o is not very different from C . Thus $\frac{\alpha' C_o^2}{\omega^2}$ may also be written as $\frac{\alpha' C_o^2}{\omega^2} = \frac{\alpha' \lambda^2}{4\pi^2}$, where λ is the wavelength of the sound wave. Moreover, in most cases $(\alpha' \lambda)^2 / 4\pi^2 \ll 1$. Under these conditions, (1-79) can be approximated by

$$\frac{C_o^2}{C^2} = \frac{1 + \omega^2 \tau_{VS} \tau_{pS}}{1 + \omega^2 \tau_{pS}^2} .$$

However, from (1-66)

$$\tau_{VS} \tau_{pS} = \tau_{pS}^2 \frac{K_S^\infty}{K_S^o} = \tau_{pS}^2 (1 - \epsilon) ,$$

so that finally

$$\frac{C_o^2}{C^2} = 1 - \frac{\epsilon \omega^2 \tau_{pS}^2}{1 + \omega^2 \tau_{pS}^2} . \quad (1-80)$$

Plotted on a logarithmic frequency scale, this function exhibits a dispersion step with an inflection point at

$$\omega = \frac{1}{\tau_{pS}} .$$

If on the other hand, the imaginary parts of (1-78) are compared, it is easily seen that

$$\frac{2 \alpha' C_o^2}{\omega C} = \frac{\alpha' \lambda}{\pi} \frac{C_o^2}{C^2} = \frac{\omega (\tau_{pS} - \tau_{VS})}{1 + \omega^2 \tau_{pS}^2}$$

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$\alpha' \lambda$ is called the molecular absorption index, and is designated by μ or α_λ . Using (1-80) a little simplification yields:

$$\mu \frac{c_o^2}{c^2} = \pi \epsilon \frac{\omega \tau_{ps}}{1 + \omega^2 \tau_{ps}^2} \quad (1-81)$$

and

$$\mu = \alpha' \lambda = \frac{\pi \epsilon}{\sqrt{1 - \epsilon}} \cdot \omega \frac{\sqrt{\tau_{ps} \tau_{vs}}}{1 + \omega^2 \tau_{ps} \tau_{vs}} \quad (1-82)$$

When plotted against $\ln \omega$, both $\mu \frac{c_o^2}{c^2}$ and μ are bell-shaped curves. The maxima of these curves are at $\omega = \frac{1}{\tau_{ps}}$

and $\omega = \frac{1}{\sqrt{\tau_{ps} \tau_{vs}}}$ respectively.

The curves of $\frac{c_o^2}{c^2}$, $\mu \frac{c_o^2}{c^2}$ and μ appear in fig. (1) a), b) and c) respectively.

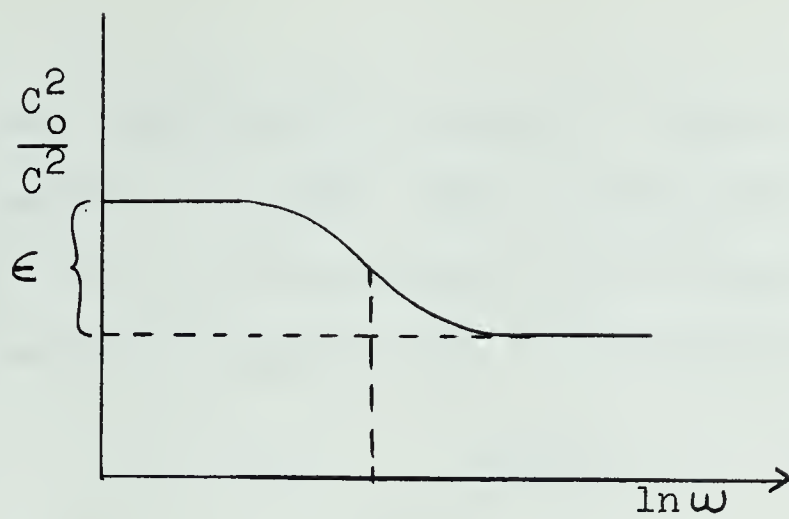
Acoustic measurements can therefore yield the values of τ_{ps} and τ_{vs} . Equation (1-81) in particular constitutes the basis for the measurements reported in this thesis; the molecular absorption index is thus seen to be related to the two adiabatic relaxation time τ_{ps} and τ_{vs} .

On the other hand the relaxation times can be calculated by means of equations (1-65). However, the thermodynamics of irreversible processes does not provide any information about the progress variables or about the affinities. Reaction

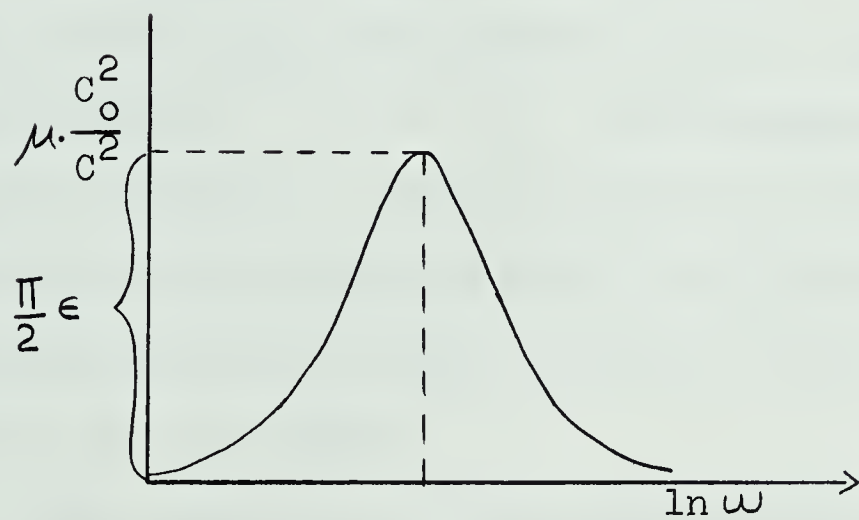
Fig. 1

- a) $(c_o/c)^2$ vs $\ln \omega$
- b) $\mu (c_o/c)^2$ vs $\ln \omega$
- c) μ vs $\ln \omega$

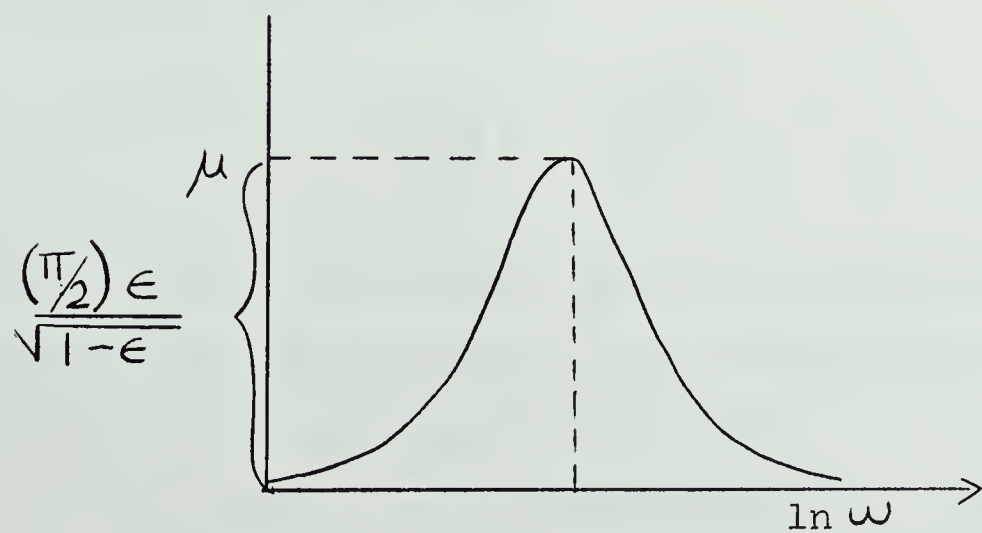
(a)



(b)



(c)





kinetics must therefore be applied in order to evaluate these parameters. For the sake of simplicity and following Kneser⁴ ζ will be calculated only for the special case where the reaction equation has the following form:



This equation describes the de-excitation and excitation of internal degrees of freedom by binary collisions. A_0 denotes the unexcited molecule and A represents the collision partner (A_0 or A_1) which remains unchanged during the collision. Although equation (1-83) is only one of the several reactions which are possible in principle, it is the only reaction which occurs as the result of collisions in a two level gas.

The population of the ground state and that of the excited state change, during the reaction to the right, according to the equation:

$$\frac{d \overline{n}_0}{dt} = - \frac{d \overline{n}_1}{dt} = R n_1$$

where n_0 and n_1 are the number of molecules per unit volume in the ground and in the excited states respectively; R is the rate constant to the right.

A similar equation describes the reaction to the left:

$$\frac{d\overleftarrow{n}_1}{dt} = - \frac{d\overleftarrow{n}_o}{dt} = R'n_o$$

Here R' is the rate constant to the left. If the molar number n_o is introduced as the progress variable ξ , the time derivative $\dot{\xi}$ includes $\frac{d\overrightarrow{n}_o}{dt}$ and $\frac{d\overleftarrow{n}_o}{dt}$. $\dot{\xi}$ can be expressed by

$$\dot{\xi} = Rn_1 - R'n_o = Rn_1 \left(1 - \frac{n_o/n_1}{R/R'} \right) \quad (1-84)$$

At equilibrium, $\dot{\xi} = 0$ and $n_{oo}/n_{1o} = R/R'$ where the additional subscript o (on the n 's) denotes the equilibrium values. At small displacements

$$\frac{n_o}{n_1} = \frac{n_{oo} + \Delta n_o}{n_{1o} + \Delta n_1} = \frac{n_{oo} + (\xi - \xi_o)}{n_{1o} - (\xi - \xi_o)} = \frac{n_{oo}}{n_{1o}} \left[1 + \left(\frac{1}{n_{oo}} + \frac{1}{n_{1o}} \right) (\xi - \xi_o) \right].$$

This value substituted into (1-84) yields

$$\dot{\xi} = - Rn_1 \left(\frac{1}{n_{oo}} + \frac{1}{n_{1o}} \right) (\xi - \xi_o) = - R \left(1 + \frac{n_{1o}}{n_{oo}} \right) (\xi - \xi_o) \quad (1-85)$$

When the experiment is conducted under isothermal conditions, ξ_o remains constant during the relaxation process. The solution of (1-85) is then

THEORY

Let $f(x)$ be a function defined on the interval $[a, b]$. The definite integral of $f(x)$ from a to b is denoted by $\int_a^b f(x) dx$. This represents the area under the curve $y = f(x)$ between $x = a$ and $x = b$.

$$\int_a^b f(x) dx = F(b) - F(a)$$

where $F(x)$ is the antiderivative of $f(x)$. The definite integral is a number, while the indefinite integral is a function.

Properties of the definite integral:

$$\int_a^b c f(x) dx = c \int_a^b f(x) dx$$

$$\int_a^b f(x) dx + \int_b^c f(x) dx = \int_a^c f(x) dx$$

The definite integral is used to calculate the area under a curve, the volume of a solid, and the average value of a function.

$$(\xi - \xi_0) = (\xi_{t=0} - \xi_0) e^{-t/\tau_T}$$

with an isothermal relaxation time given by

$$\tau_T^{-1} = R \left(1 + \frac{n_{10}}{n_{00}}\right) = (R + R') \quad . \quad (1-86)$$

When the reaction represents the excitation or the de-excitation of a molecule, the rate constants R and R' are simply proportional to the probabilities f_{01} and f_{10} :

$$R = \tau_c^{-1} f_{10} \quad , \quad R' = \tau_c^{-1} f_{01}$$

where τ_c is the mean collision time; f_{10} is the probability that an excited molecule will return to the ground state when colliding with another one, while f_{01} is the probability that a molecule in the ground state will be excited when colliding with another molecule. Equation (1-86) shows that the isothermal relaxation time is inversely proportional to the sum of the rate constants.

The experimental values of the sound absorption or of the velocity dispersion yield the adiabatic relaxation times τ_{pS} and τ_{vS} , while reaction kinetics calculations yield the isothermal relaxation times τ_{pT} or τ_{vT} . In order to compare the experimental values with the theoretical

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values, the former must be converted into isothermal values by means of equations (1-67) and (1-68). The difference between the isothermal and the adiabatic relaxation times is often minimal, especially in the case of vibrational relaxation. It is not so however in the case of rotational relaxation. The various relaxation times are clearly indicated on fig. (2).

The following inequalities always hold:

$$\tau_{pT} > \tau_{pS} > \tau_{VS} \quad .$$

This distinction between the various relaxation times has often been ignored in the literature. Nowhere has it been expressed more clearly however, than in the papers by Bauer¹ and by Kneser.⁴

4. Relaxational Absorption in Hydrogen.

In order to compare experimental results with theoretical calculations, it is best to effect measurements on a simple molecule. Limiting ourselves to the problem of rotational energy transfer, the hydrogen molecule at low temperature constitutes such a simple model for the following reasons:

The first part of the paper discusses the importance of the study of the history of the English language. It is argued that the study of the history of the English language is not only a matter of historical interest, but also a matter of practical importance. The study of the history of the English language is essential for the understanding of the English language in its present state. The study of the history of the English language is also essential for the understanding of the English language in its future state. The study of the history of the English language is also essential for the understanding of the English language in its present and future state.

THE HISTORY OF THE ENGLISH LANGUAGE

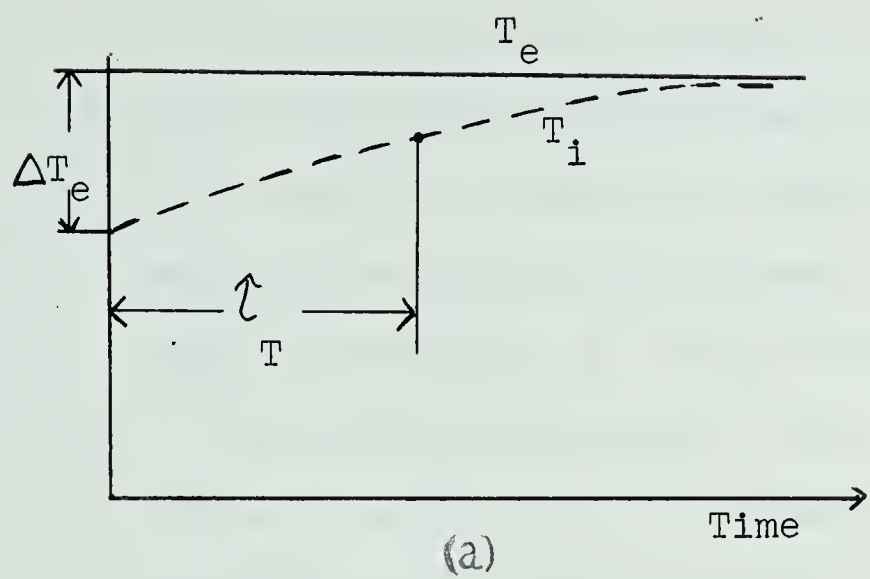
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Fig. 2

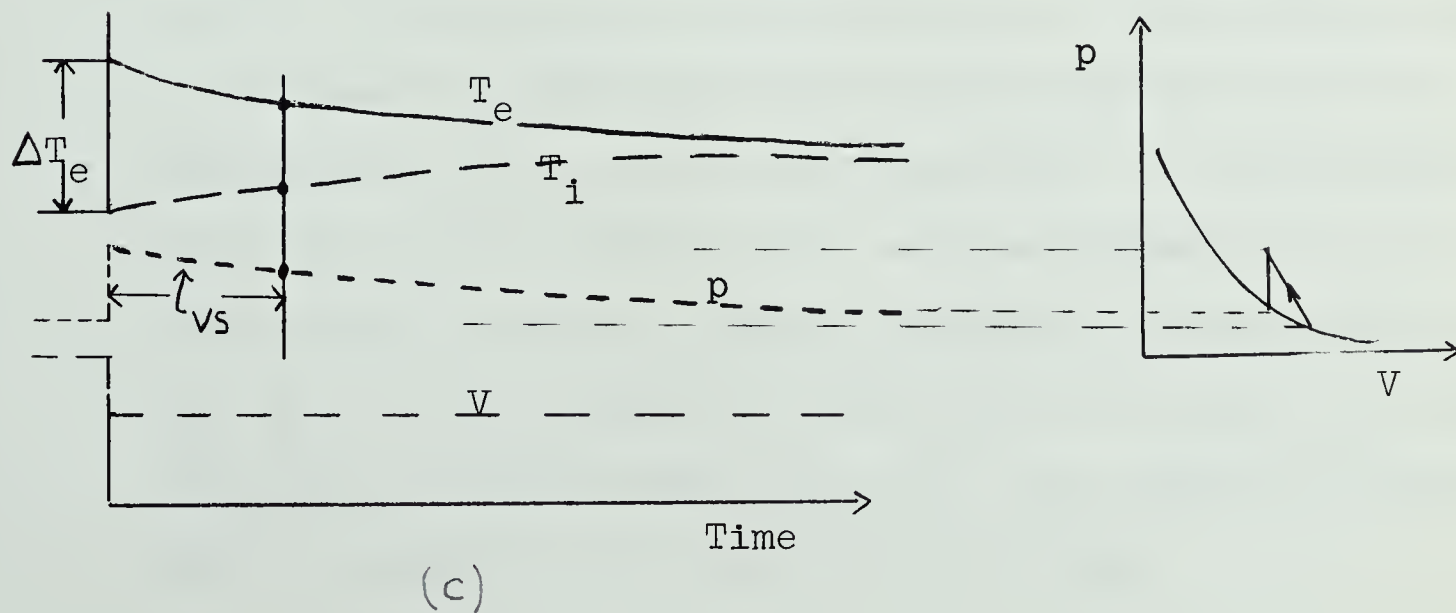
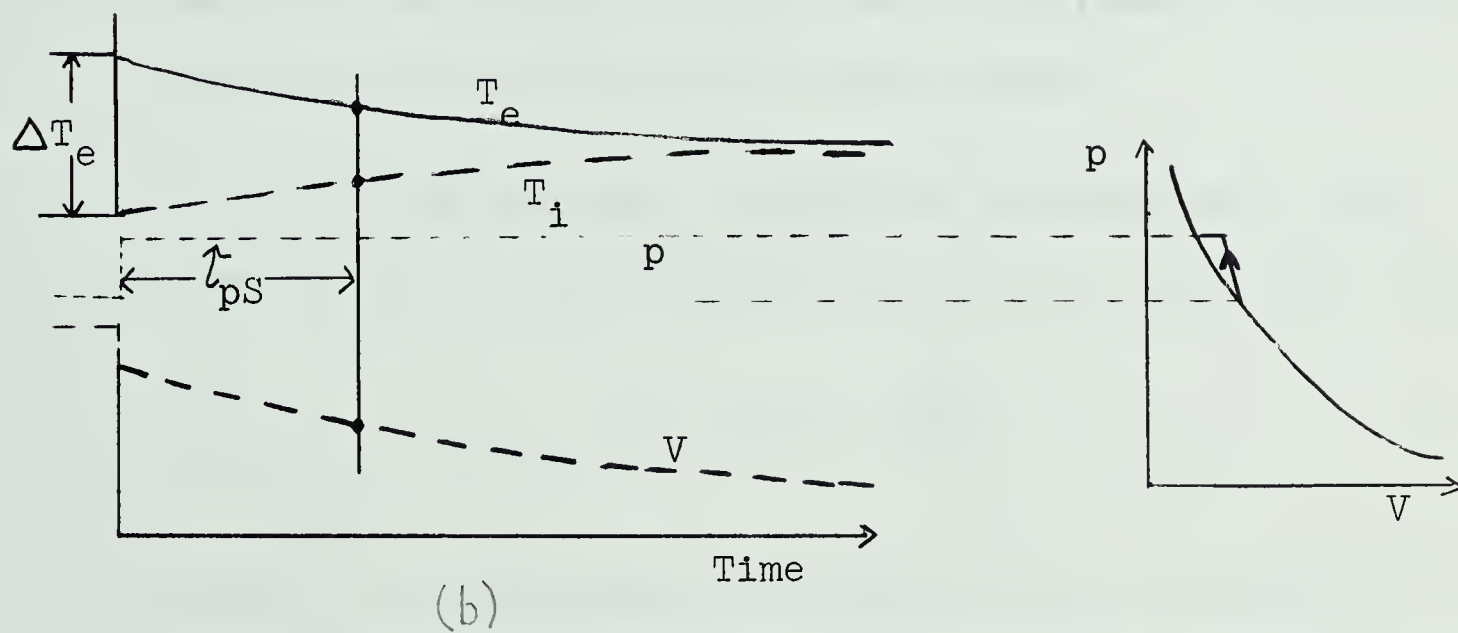
- a) τ_{pT}
- b) τ_{pS}
- c) τ_{VS}



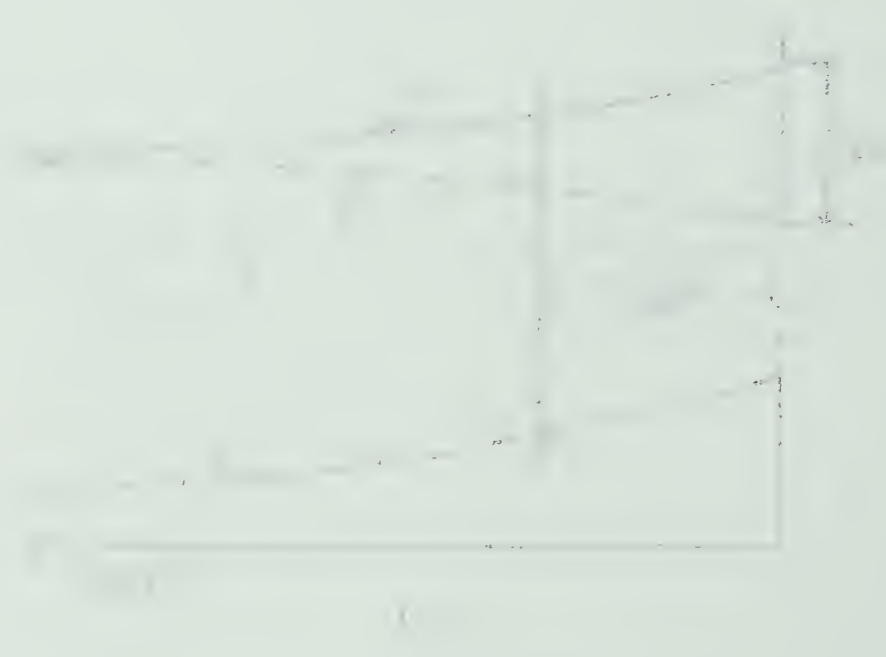


T_e : external temperature

T_i : internal temperature



1. $\frac{1}{x^2} = x^{-2}$
 $\frac{d}{dx} x^{-2} = -2x^{-3} = -\frac{2}{x^3}$



1) depending on the isotopic constitution of the molecules, only two levels are important at temperatures ranging from 20 to 150° K -- when more than two levels are occupied, multiple relaxation processes occur, which are rather difficult to analyse; 2) due to the very small moment of inertia of the hydrogen molecule, the rotational energy levels are widely spaced; 3) the relaxation strength ϵ can be computed easily since physical properties such as the specific heats are well known in the case of hydrogen. These three points will now be examined more closely.

The hydrogen molecule is essentially a rigid rotator whose energy levels are given by the usual formula

$$E_J = J(J + 1) \frac{\hbar^2}{2I}$$

where I is the moment of inertia of the rotator, J is the rotational quantum number, \hbar is Planck's constant divided by 2π . Depending on the orientation of the nuclear spins of the two hydrogen atoms which constitute the molecule, hydrogen can exist in two modifications: orthohydrogen when the total nuclear spin is one, and parahydrogen when the total nuclear spin is zero. Quantum mechanical considerations show that only the odd-numbered rotational states can be occupied in the case of orthohydrogen, and only even-numbered rotational levels can be occupied in the case of parahydrogen.

The first part of the paper is devoted to a discussion of the
general principles of the theory of the structure of the
crystal lattice. It is shown that the structure of the
crystal lattice is determined by the arrangement of the
atoms in space. The arrangement of the atoms in space
is determined by the forces of attraction and repulsion
between the atoms. The forces of attraction and repulsion
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$$\sum_{i=1}^n \frac{1}{i^2} = \frac{\pi^2}{6}$$

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The thermodynamical equilibrium between the ortho and the para modifications is given by Boltzmann's distribution law, and simple calculations show that at room temperature, orthohydrogen ($o\text{-H}_2$) represents 75% of the equilibrium mixture. At 20°K however, the equilibrium mixture is over 99% parahydrogen ($p\text{-H}_2$). Transitions between the two modifications do not occur except in the presence of a catalyst. For example, in the absence of a catalyst, normal hydrogen (75% ortho - 25% para) does not convert to pure parahydrogen, even when cooled down to 20°K . The relative concentration of ortho and para molecules, 3:1 remains the same. As a result, the ortho and para modifications can be treated as two different gases. In the presence of a catalyst however, almost pure parahydrogen can be obtained by cooling hydrogen down to 20°K .

The occupation of the rotational energy levels as a function of temperature for $p\text{-H}_2$ and $o\text{-H}_2$ appears in figure (3) a) and b) respectively. As can be seen in these figures, below 100°K , parahydrogen is a two-level gas, which can therefore exhibit a single relaxation process, under the influence of a sound wave.

The fact that the hydrogen molecule has a small moment of inertia is of great importance; indeed, the spacing between the rotational energy levels of a molecule is

Fig. 3 a)

Energy level population in parahydrogen.

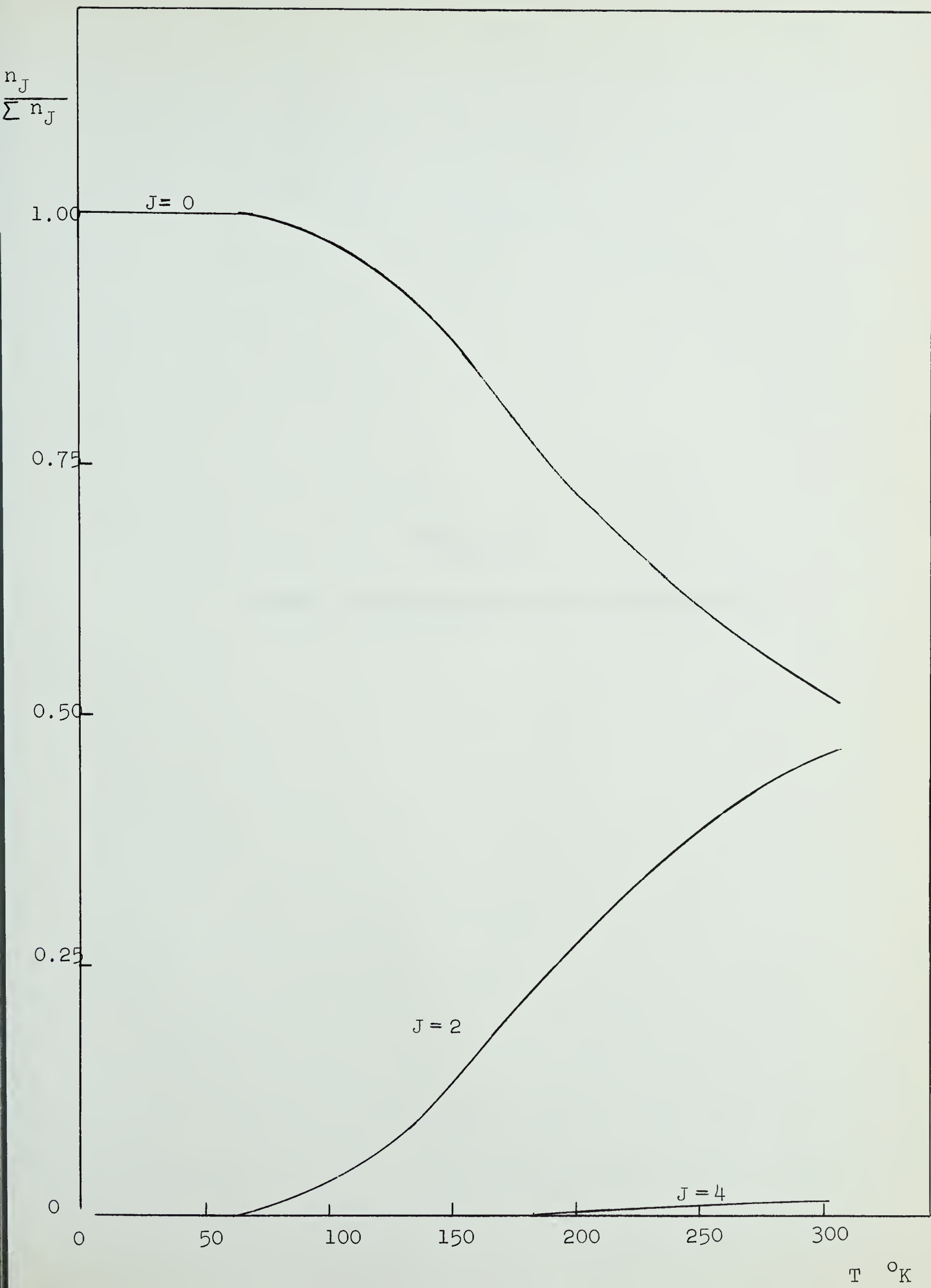
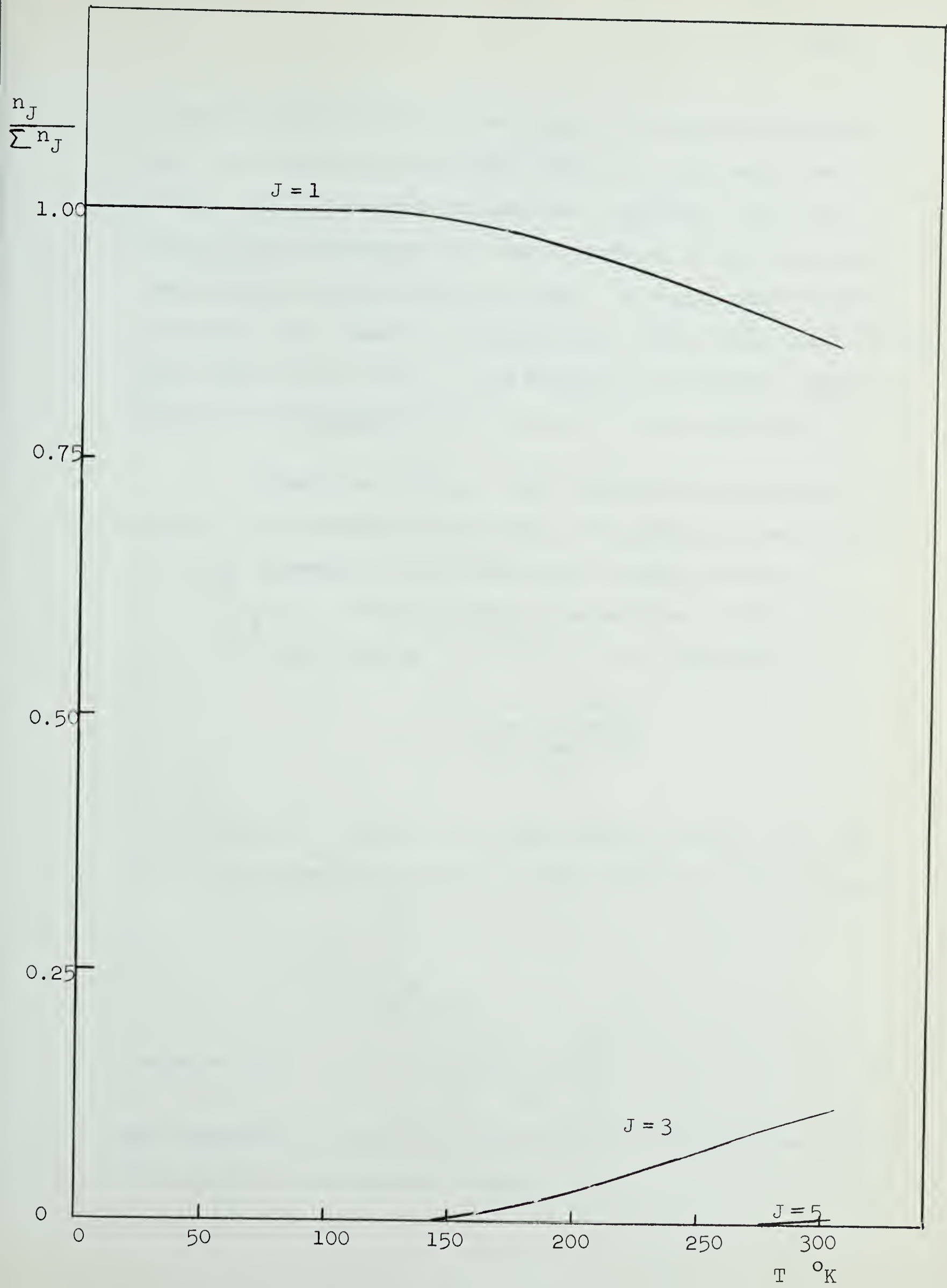


Fig. 3 b)

Energy level population in orthohydrogen.





inversely proportional to the moment of inertia of the molecule, and transitions are less likely to occur when the energy levels are widely separated. Therefore, one can conclude that molecules with small moments of inertia have comparatively long relaxation times. In fact, most molecules have such large moments of inertia that their rotational relaxation times are too short to be measured accurately: hydrogen, deuterium and hydrogen deuteride are a few exceptions.

Equations (1-80), (1-81) and (1-82) contain the factor ϵ , the relaxation strength. As mentioned previously, ϵ is the diameter of the "Cole plot", and is equal to $(1 - \tau_{VS}/\tau_{PS})$. With the help of equations (1-66), (1-67) and (1-68), and putting $K_T^0/K_T^\infty = 1$, one can write

$$\epsilon = \frac{C_V^0 C_p^\infty - C_V^\infty C_p^0}{C_V^0 C_p^\infty}$$

Introducing C' as the rotational specific heat, i.e. that part of the specific heat due to the rotation of the molecule,

$$C_p^\infty = C_p^0 - C'$$

and

$$C_V^\infty = C_V^0 - C'.$$

Consequently,

$$\epsilon = \frac{C'(C_p^0 - C_V^0)}{C_V^0 (C_p^0 - C')}.$$

The relaxation strength is thus seen to be directly related to the values of the specific heats.

CHAPTER II

EXPERIMENT

1. Experimental Methods

The methods which have been devised for measuring the absorption and the dispersion of sound waves, can be classified into three groups: 1) mechanical, 2) optical, and 3) electrical. In one section of a comprehensive paper on the absorption of sound in fluids, Markham, Beyer and Lindsay⁵ made a critical examination of these methods, and the following paragraphs are based on this paper.

Mechanical methods make use of the radiation pressure which is the difference, due to the sound beam, between the pressure on one side of a rigid wall, and the pressure on the other side of the wall, in the medium at rest. The forces being measured are very small, and often of the same order of magnitude as other extraneous forces such as the surface tension. Cavitation effects on the other hand, set an upper limit to the sound intensity. Other factors such as reflections from the walls of the container and divergence of the beam at lower frequencies limit the usefulness and the accuracy of this method. Above 10 Mc/sec, the accuracy is $\pm 5\%$, while below 3 Mc/sec, hardly any results obtained by this method, can be relied upon.

THE
JOURNAL

OF THE
ROYAL SOCIETY

OF LONDON

IN THE
MONTH OF
JANUARY
1701

At a publick sitting of the
Royal Society, on the 12th of
January 1701, the following
papers were presented and
read, viz.

1. A paper of Mr. Christopher Wren, containing
an account of the manner of
the late great fire, and of the
damages done thereby.

2. A paper of Mr. Christopher Wren, containing
an account of the manner of
the late great fire, and of the
damages done thereby.

3. A paper of Mr. Christopher Wren, containing
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damages done thereby.

Optical methods are based on the fact that when a gas is subjected to a sound wave, the alternating regions of compression and rarefaction act as diffracting elements for a beam of light at right angles to the sound path. The sound beam transforms the gas into a diffraction grating. It is easy to see that such a method imposes stringent experimental requirements: alignment for instance is very critical. Divergence of the sound beam sets a lower limit to the range of frequencies at which experiments can be conducted. Optical methods, like mechanical methods are most trustworthy above 10 Mc/sec.

The majority of measurements on sound absorption and dispersion in gases have been obtained by electrical methods, which include all methods making use of a microphone. Although piezoelectric transducers are the commonest type of microphones, transducers of the condenser type with solid dielectric have proved to be equally reliable and more versatile. Electrical methods can in turn be classified into three groups: a) interferometric, b) direct, and c) pulse.

The first ultrasonic interferometer was built by Pierce in 1925. Shortly after, Hubbard made significant contributions to the field of ultrasonic interferometry. Most of the earlier results on the absorption and dispersion of sound in gases were obtained by this method. An acoustic interferometer is essentially an electromechanical system

composed of a column of fluid set into longitudinal vibrations at one end by an oscillating plate, and terminated at the other end by a reflecting plate. Multiple reflection takes place between the source and the reflector; as the length of the sound path is varied, cyclical changes of phase and amplitude of the forced vibrations occur, the reflector passing through successive positions of resonance in the fluid medium. Due to the nature of the transducer, the variations of pressure at the surface of the vibrator manifest themselves as variations of current, or sometimes of frequency, in the electric circuit coupled to the transducer. Since it is essentially a problem of standing waves, these cyclical changes occur each time the reflector is moved by half a wavelength. The interferometer thus lends itself most conveniently to the measurement of the sound velocity. At first glance, it would appear that the absorption coefficient could be readily obtained from the ratio of the amplitude of two successive maxima. In practice however, many factors such as the reflection coefficient of the reflector have to be taken into account when calculating the absorption coefficient.

Pumper⁶ criticized this method and demonstrated that the interferometer can be made to yield differing results by slight modifications of technique. The scatter of the

The first part of the report deals with the general situation of the country and the progress of the work during the year. It is followed by a detailed account of the various projects and the results achieved. The report concludes with a summary of the work done and the prospects for the future.

The second part of the report contains a list of the names of the persons who have been engaged in the work during the year. It also contains a list of the names of the persons who have been engaged in the work during the year.

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The report is signed by the Director of the Institution, who is also the President of the Council of the Institution. It is dated the 1st day of January, 1900.

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results obtained by this method for the values of the sound absorption coefficients seem to justify Pumper's criticisms. As stated previously however, the interferometer remains most useful and reliable for measuring the sound dispersion.

In the direct method, a transducer of radius a generates a sound wave of wavelength λ . Within a distance a^2/λ from the emitter, Fresnel diffraction takes place, and the sound beam is cylindrical with a radius a . Within that distance, the amplitude of the sound beam averaged over a cross-section of the cylinder decreases as $e^{-\alpha x}$, where α is the absorption coefficient of the medium, and x is the distance from the source. Standing waves are a problem, although at high enough absorption and at large distances, their effect becomes negligible. Mechanical and electrical cross-talks also give difficulties if they are not neutralized properly. In gases, this method has been applied quite successfully at 3 Mc/sec/atm and higher, in particular, by Meyer and Sessler⁷, and by Sluijter, Knaap and Beenakker.⁸

All methods making use of continuous waves present the following disadvantages: standing waves can affect the results, and moreover, if power is continuously dissipated in the form of heat in the medium, the temperature of the fluid may be altered locally, and measurements taken at a temperature higher than that recorded. In the pulse method,

these problems do not arise: if the pulse repetition rate is low enough, standing waves cannot be set up, and if the duty ratio is of the order of 1:500, the heating effect is reduced in the same proportion. The pulse method also permits to distinguish in time, between the received pulse and other unwanted signals. One objection may be raised against the pulse method however; since a short pulse contains a whole spectrum of frequencies, and since the sound absorption varies with the frequency, there is an error involved in taking measurements with short pulses. Pellam and Galt⁹ demonstrated that when the absorption coefficient varies as the square of the frequency, and when the pulse contains several oscillations, the error involved is equal to the square of the reciprocal of the number of oscillations per pulse. For example, when a pulse contains ten oscillations, the error is one part in a hundred, which is usually less than the errors associated with the measurement of other parameters, such as the pressure, the voltage etc. The main disadvantage of the pulse method is the poor signal-to-noise ratio associated with the large bandwidth of the receiver.

In conclusion , no method is obviously the best one. A compromise must be reached, which depends on the problem to be solved, as well as on the facilities available in a particular laboratory.

The first part of the book is devoted to a study of the
history of the Church in the Middle Ages. The author
discusses the development of the Church from the time of
the apostles to the present day.

The second part of the book is devoted to a study of the
doctrines of the Church. The author discusses the development
of the Church's teaching from the time of the apostles to the
present day.

The third part of the book is devoted to a study of the
liturgy of the Church. The author discusses the development
of the Church's worship from the time of the apostles to the
present day.

The fourth part of the book is devoted to a study of the
pastoral work of the Church. The author discusses the development
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present day.

The fifth part of the book is devoted to a study of the
social work of the Church. The author discusses the development
of the Church's social teaching from the time of the apostles to the
present day.

The sixth part of the book is devoted to a study of the
missionary work of the Church. The author discusses the development
of the Church's mission from the time of the apostles to the
present day.

The seventh part of the book is devoted to a study of the
ecumenical work of the Church. The author discusses the development
of the Church's ecumenical teaching from the time of the apostles to the
present day.

The eighth part of the book is devoted to a study of the
future of the Church. The author discusses the development
of the Church's future from the time of the apostles to the
present day.

2. Apparatus

In order to observe a single relaxation process in hydrogen, measurements have to be taken at temperatures below 150°K. Since liquid nitrogen was the most readily available refrigerant, a gas vessel was constructed which is suitable for that temperature. Another requirement imposed on the apparatus was the range of frequency-to-pressure ratios (f/p) at which experiments were to be conducted. Since the maximum of the absorption peak for hydrogen occurs at around 10 Mc/sec/atm, the apparatus was built to cover an appropriate range of f/p : from 3 to 50 Mc/sec/atm. Equations (1-80), (1-81) and (1-82) show that τ always appears in the combination $\omega\tau$. Assuming that rotational energy transfer is due to binary collisions only, τ is inversely proportional to the collision rate, and consequently inversely proportional to the pressure. The absorption can thus be measured as a function of f/p , rather than of $\omega\tau$. The proportionality factor between f/p and $\omega\tau$ is independent of frequency and pressure. Since it is easier experimentally to vary the pressure rather than the frequency, measurements were taken at a fixed frequency, for various pressure values.

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27. The twenty-seventh part of the document is a list of the names of the members of the committee.

28. The twenty-eighth part of the document is a list of the names of the members of the committee.

A. Mechanical part.

A schematic diagram of the cryostat appears in fig. 4. The cryostat is supported vertically and can thus be immersed in a refrigerant bath. The double bellows arrangement, adapted from Stewart,¹⁰ has two advantages: firstly, the volume remains constant while the distance between the two transducers is varied, and secondly, this arrangement constitutes a temperature-resistant motion seal -- other types of motion seals while efficient at room temperature, may not be leak-proof at liquid nitrogen temperature. The elasticity of the bellows permits the length of the sound path to be varied from 1.5 to 14 mm. The position of the movable transducer is determined by means of a micrometer.

Indium O-rings are used as seals between the stainless steel flanges which are soldered to the bellows, and the flanges of the central rigid portion of the cryostat.

Spring loaded adjustment screws permit the transducers to be set as parallel as possible. The right degree of parallelism is achieved when the received signal is maximum. It is also important that this parallelism be maintained as the movable transducer is brought forward or backward: This is the purpose of the driving shaft fixed to the upper plate of the cryostat, which slides inside a tight-fitting bushing.

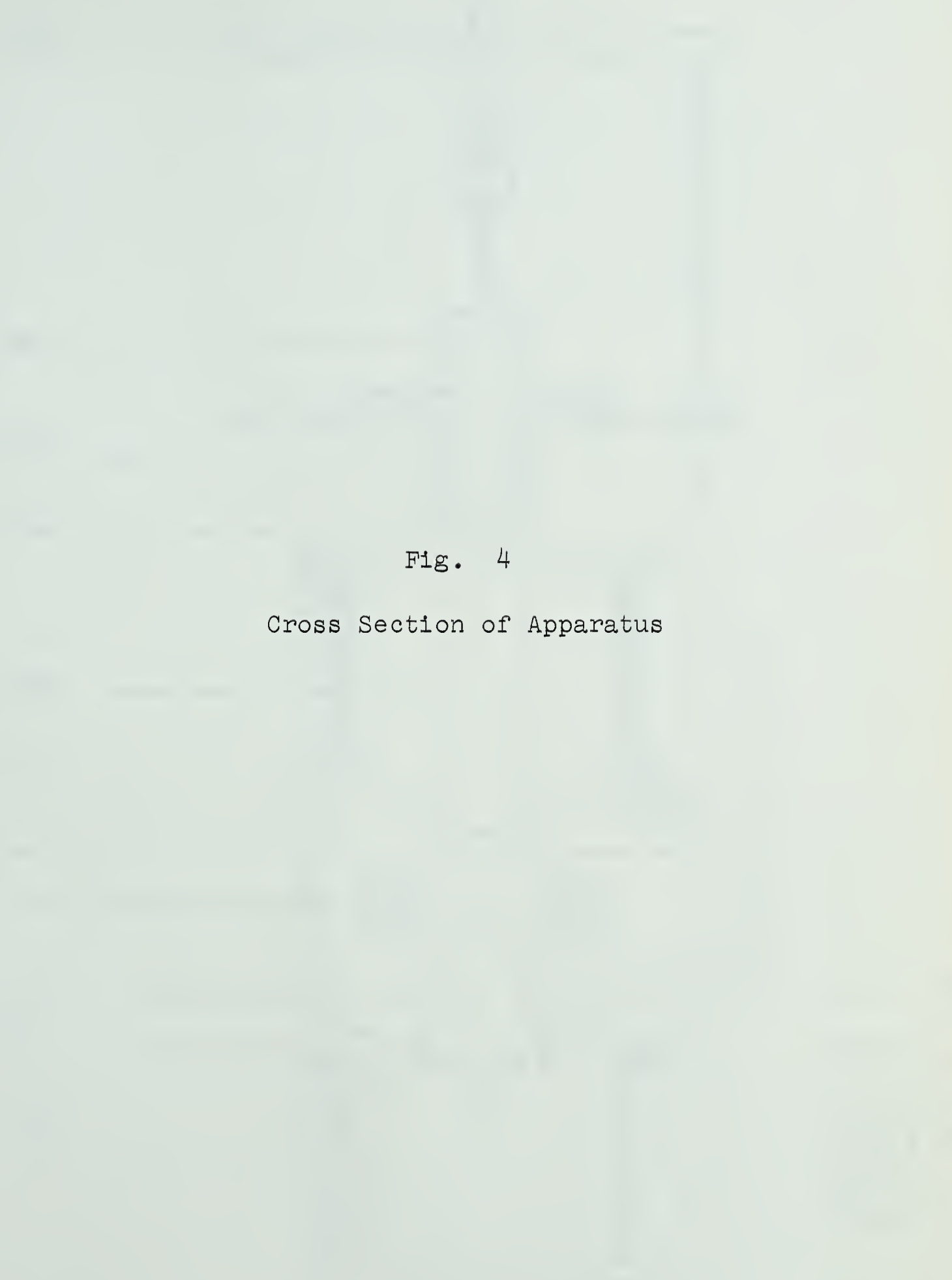
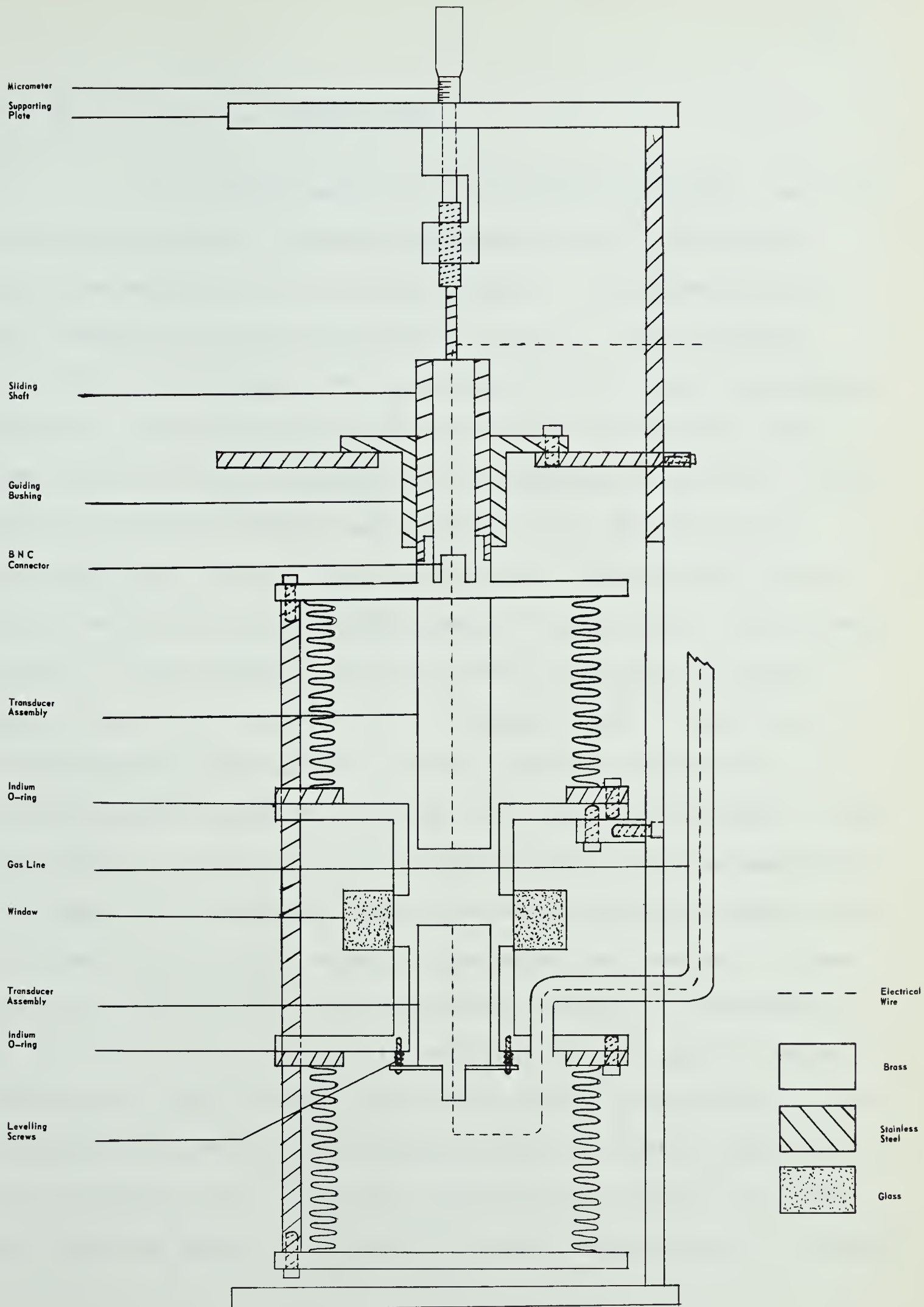


Fig. 4

Cross Section of Apparatus

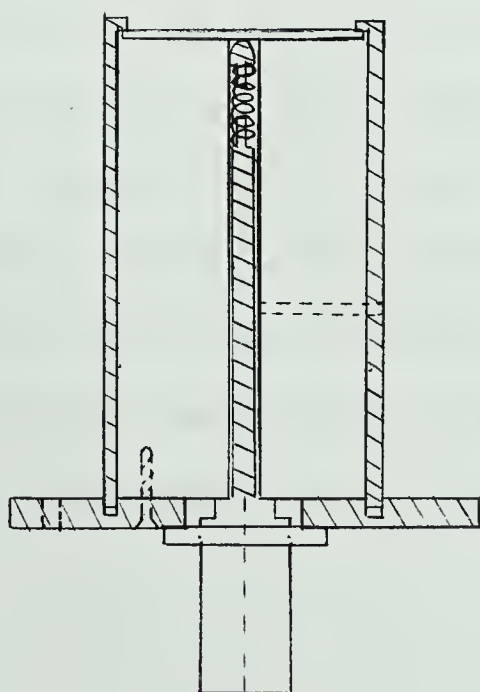
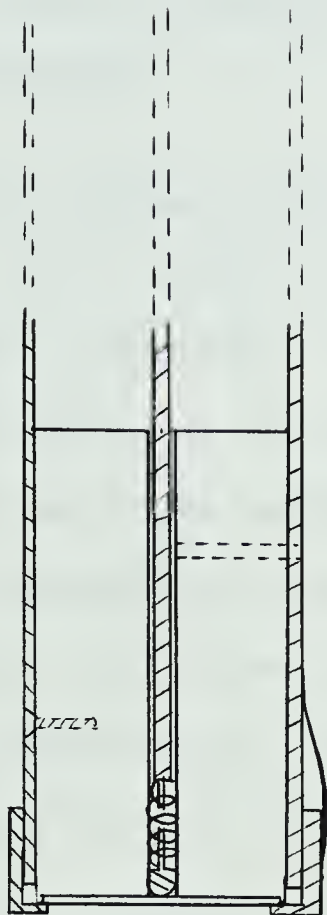


B. Mounting of the Transducers

The transducers are X-cut quartz crystals, one inch in diameter, with a fundamental frequency of 3.00 Mc/sec. They are cemented to a block of Lucite. A cross-section of the transducers support appears in fig. 5. Many attempts were made at providing the transducers with proper electrodes. Films of copper, aluminum and chromium were deposited onto the surfaces of the transducers by evaporation in vacuo. The electrodes thus obtained were too thin and did not adhere satisfactorily to the quartz substrate. Coating the transducers with silver metallizing paint (electrically conducting), proved to be successful to some extent, although the paint had a tendency to dissolve in the grease used to bond the transducers to the Lucite. Another attempt consisted of cementing thin discs of aluminum foil onto the surfaces of the transducers. This also was a rather satisfactory arrangement, although the transducers seemed to be too heavily loaded, with the result that their resonant frequency was lowered. Consequently, the following compromise was reached: the surface of the transducer which is in contact with the gas is coated with silver paint, while the surface which is cemented to the Lucite is covered with an electrode cut out from a sheet of thin aluminum foil. Silicone vacuum grease proved to be the best bonding agent. At liquid nitrogen temperature, the resonant

Fig. 5
Transducers Mount





frequency of the transducers is 2.85 Mc/sec. The lowering of the resonant frequency is seemingly due to the extra thickness of the foil and the thin film of grease used to bond the electrode to the quartz crystal.

Unloaded quartz crystals have a very high "Q" factor, which can reach 10,000. Such a highly tuned device cannot respond to a pulse of short duration. In the pulse method, it is thus necessary to reduce the "Q" factor of the transducer, or in other words, to increase its bandwidth. This is achieved by backing the transducer with a high impedance and high loss material. Lucite satisfies these two conditions, and is used to load the transducers. Cemented to a block of Lucite, the quartz crystals have a "Q" of approximately 15.

Electromagnetic shielding of transducers calls for coaxial types of connectors. It is for this reason that the arrangement shown in fig. 5 was adopted: the ground connection is provided by a brass hollow cylinder surrounding the cylindrical block of Lucite through the centre of which passes the "live" connector - a thin brass rod terminated in a spring loaded plug. One end of the brass cylinder is such that it covers the periphery of the transducer's face, thus providing electrical contact to the ground.

The first of these is the fact that the system is not a simple one. It is a complex system, and the complexity is not only in the number of variables, but also in the nature of the variables. The second is the fact that the system is not a simple one. It is a complex system, and the complexity is not only in the number of variables, but also in the nature of the variables.

The third is the fact that the system is not a simple one. It is a complex system, and the complexity is not only in the number of variables, but also in the nature of the variables. The fourth is the fact that the system is not a simple one. It is a complex system, and the complexity is not only in the number of variables, but also in the nature of the variables. The fifth is the fact that the system is not a simple one. It is a complex system, and the complexity is not only in the number of variables, but also in the nature of the variables.

The sixth is the fact that the system is not a simple one. It is a complex system, and the complexity is not only in the number of variables, but also in the nature of the variables. The seventh is the fact that the system is not a simple one. It is a complex system, and the complexity is not only in the number of variables, but also in the nature of the variables. The eighth is the fact that the system is not a simple one. It is a complex system, and the complexity is not only in the number of variables, but also in the nature of the variables.

C. Vacuum system

The gas vessel is evacuated by means of a mechanical pump capable of an ultimate vacuum of 10^{-3} mm Hg. As a preliminary measure, the apparatus was checked for leaks, and connected to a mercury diffusion pump. After evacuation by means of the diffusion pump, the vessel maintains overnight a vacuum better than 10^{-3} mm Hg.

Pressures are read from a mercury manometer with an accuracy of ± 0.5 mm.

D. Electronic Circuitry

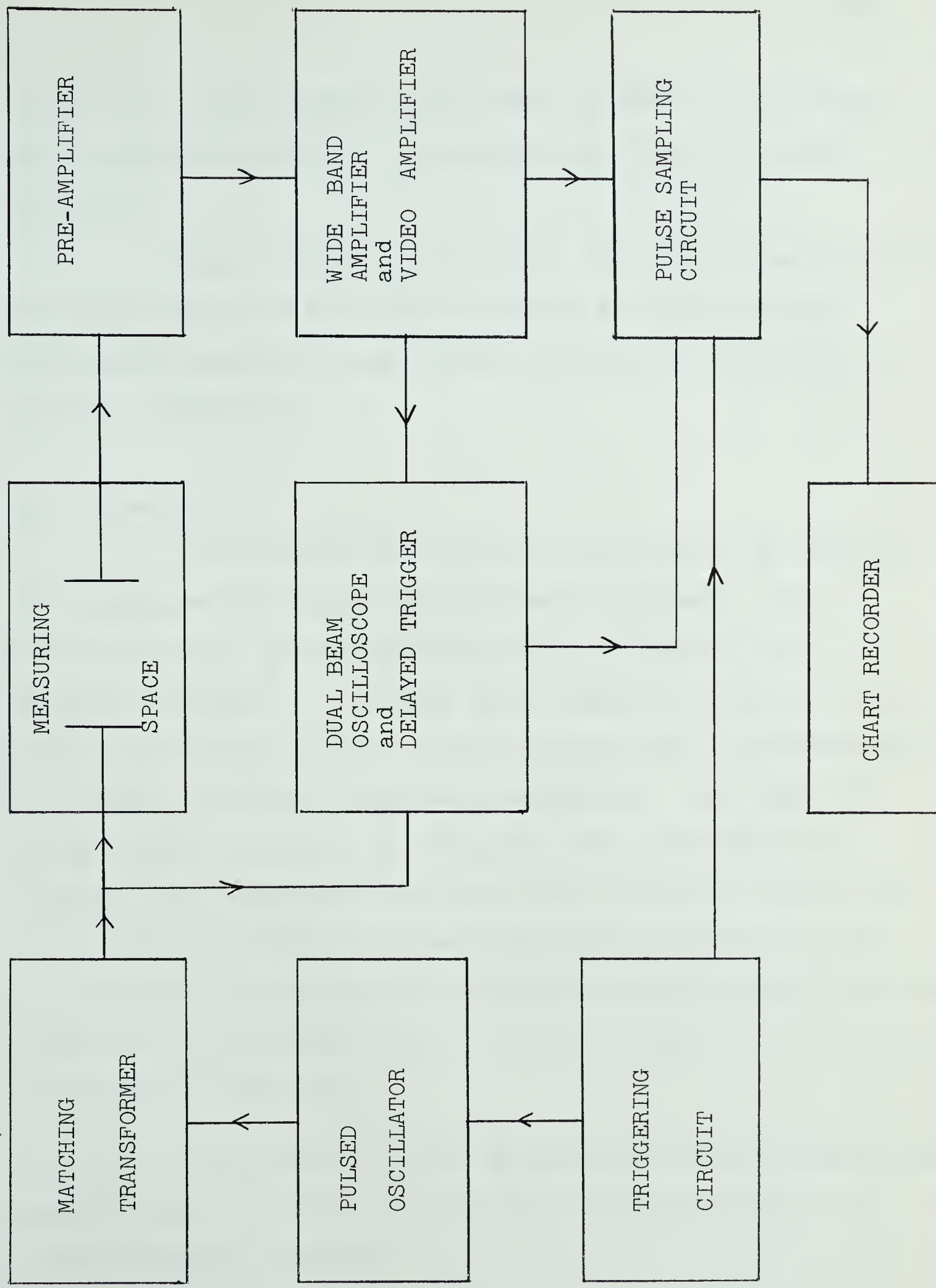
A block diagram of the electronic components appears in fig. 6.

A R.F. pulse of adjustable duration, frequency and repetition rate originates from an oscillator driven by a blocking oscillator. Matching of the transducer to the output of the oscillator is effected by means of a tuned air-core transformer with variable number of turns and variable turns ratio.

The receiving transducer is connected to a preamplifier with tunable input coils. The signal is then amplified in a wide-band amplifier with a maximum gain of 80 db. The video output signal from the amplifier is measured by means of a

Fig. 6

Electronic Circuitry





"sampling" circuit adapted from Kamm and Bohm.¹¹ The output of the sampling circuit is recorded on a 2.5 mv full-scale detector.

Such a method requires that the gain of the amplifier be very stable; for this, all filaments of the amplifying system were heated by large capacity batteries connected to a battery eliminator.

3. Procedure

The cryostat is filled with the gas to be examined, to a pressure slightly above atmospheric pressure. The received signal is recorded at various positions of the movable transducer. A typical record appears in fig. 7. Each record contains in general fifteen double peaks, corresponding to fifteen different transducers separation. The peaks are double since each pulse is "sampled" twice, backward and forward. The amplitudes are then read from these records and the natural logarithm of these amplitudes are plotted versus the distance. The absorption coefficient is determined from the slope of these straight lines. A typical graph of $\ln A$ versus x is shown in fig. 8.

The pressure is then decreased to the next lower value, and the above procedure is repeated, similarly down to the lowest workable pressure.

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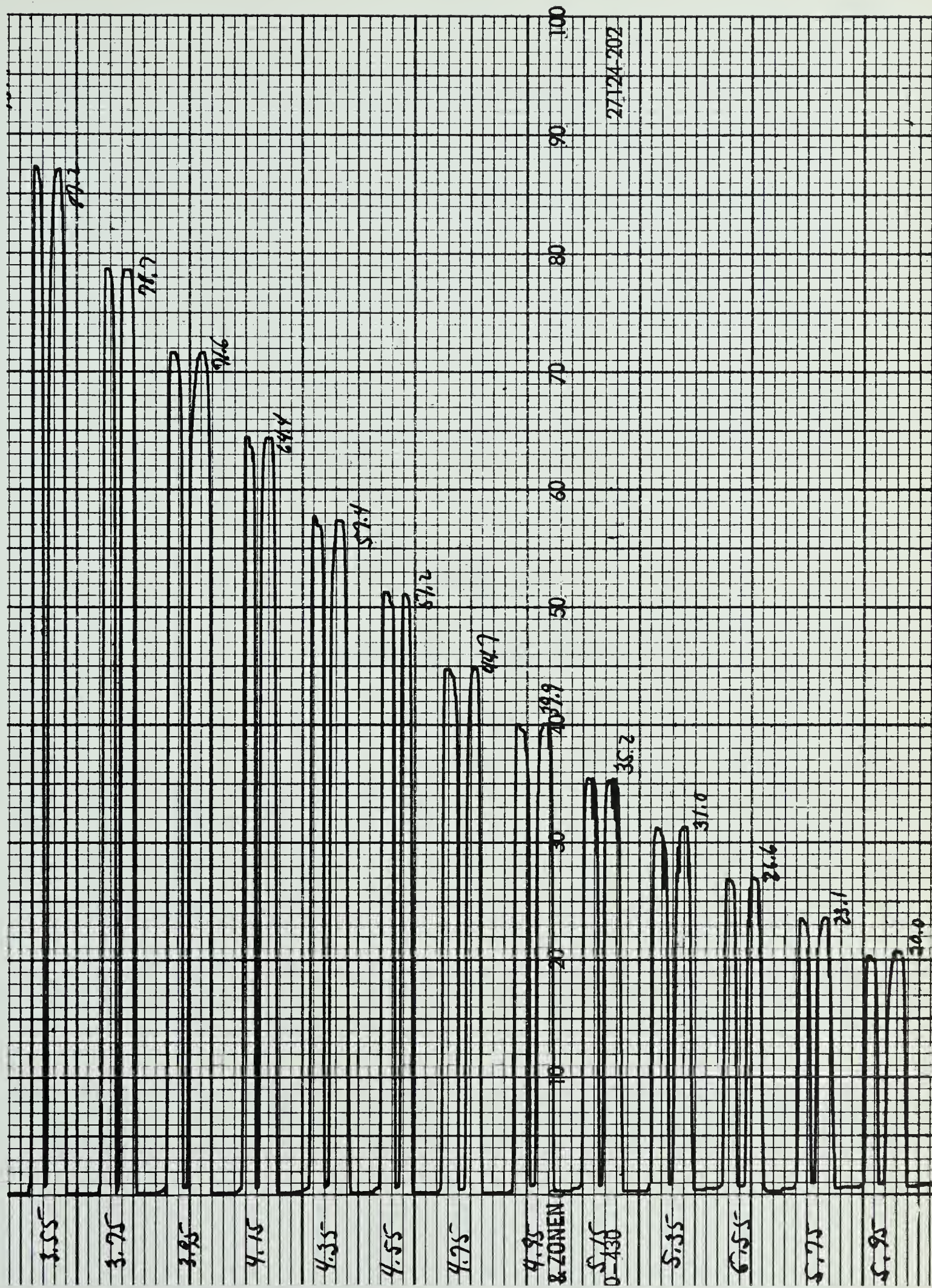
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Fig. 7

Typical Record



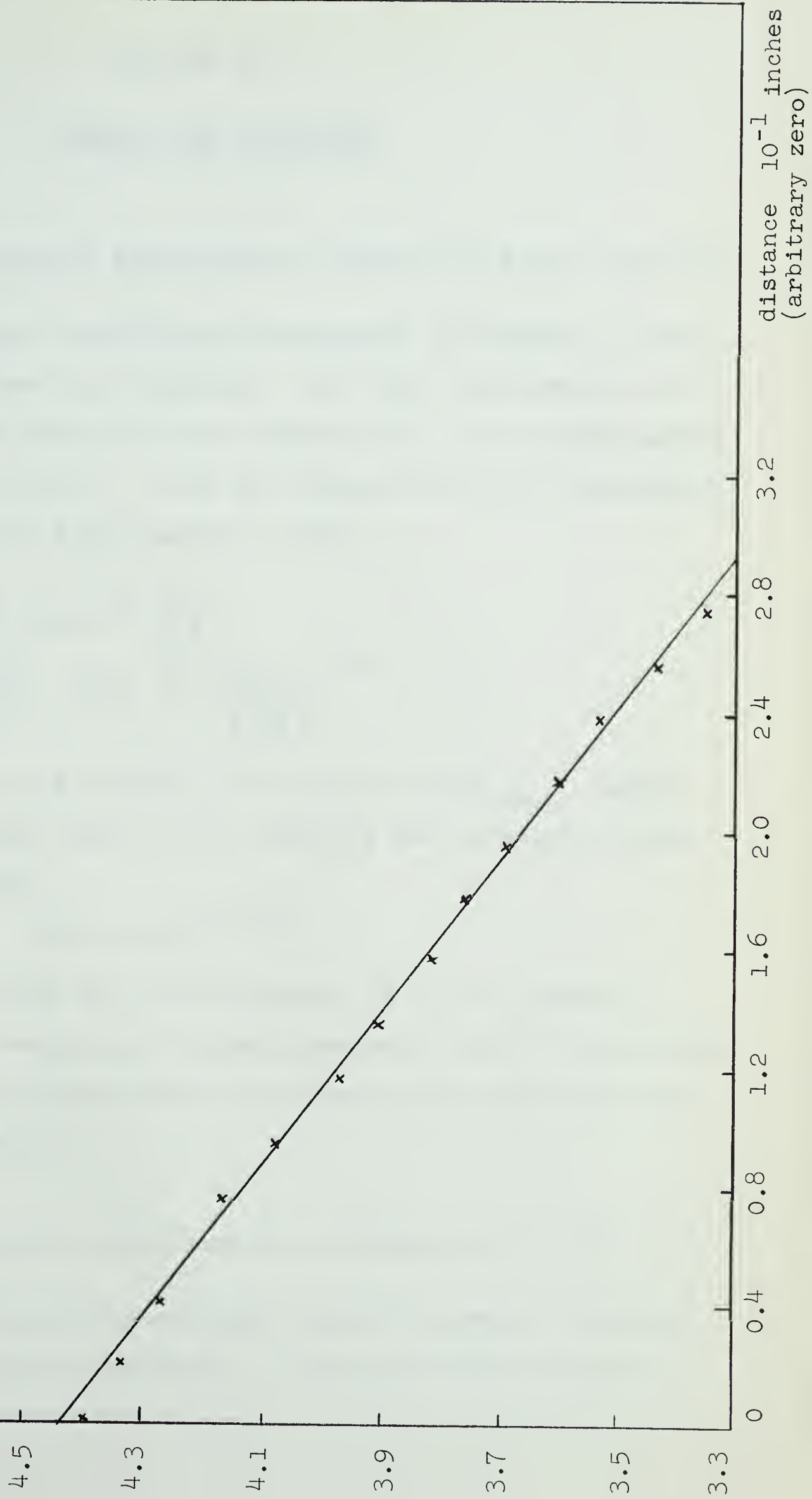
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No.		Date		Description		Amount		Total	
1		Jan 1		Balance		100		100	
2		Jan 10		Received		50		150	
3		Jan 20		Received		25		175	
4		Jan 30		Received		25		200	
5		Feb 10		Received		50		250	
6		Feb 20		Received		25		275	
7		Feb 30		Received		25		300	
8		Mar 10		Received		50		350	
9		Mar 20		Received		25		375	
10		Mar 30		Received		25		400	
11		Apr 10		Received		50		450	
12		Apr 20		Received		25		475	
13		Apr 30		Received		25		500	
14		May 10		Received		50		550	
15		May 20		Received		25		575	
16		May 30		Received		25		600	
17		Jun 10		Received		50		650	
18		Jun 20		Received		25		675	
19		Jun 30		Received		25		700	
20		Jul 10		Received		50		750	
21		Jul 20		Received		25		775	
22		Jul 30		Received		25		800	
23		Aug 10		Received		50		850	
24		Aug 20		Received		25		875	
25		Aug 30		Received		25		900	
26		Sep 10		Received		50		950	
27		Sep 20		Received		25		975	
28		Sep 30		Received		25		1000	
29		Oct 10		Received		50		1050	
30		Oct 20		Received		25		1075	
31		Oct 30		Received		25		1100	
32		Nov 10		Received		50		1150	
33		Nov 20		Received		25		1175	
34		Nov 30		Received		25		1200	
35		Dec 10		Received		50		1250	
36		Dec 20		Received		25		1275	
37		Dec 30		Received		25		1300	
38		Jan 1		Balance		1300		1300	

Fig. 8

Curve of $\ln A$ vs x

n-H₂
1/p : 2.5 atm⁻¹
f/p : 7.12 Mc/sec/atm
 α : 1.56 cm





CHAPTER III

RESULTS AND DISCUSSION

1. Sound Absorption Measurements in Helium at Room Temperature

Before undertaking measurements in hydrogen, it was necessary to test the apparatus. For this, measurements were first taken in helium at room temperature. The results appear in fig. 9 and table 2. They are compared with the theoretical curve calculated from equation (1-58)

$$\alpha_{\text{class}} = \frac{7}{6} \frac{1}{R}$$

where $R = \frac{\rho_o v_o^2}{\eta \omega}$, and $v_o = \left(\frac{\gamma p_o}{\rho_o} \right)^{1/2}$.

The data necessary for calculating α_{class} appear in table 1. The value of the viscosity was calculated from Keesom's formula

$$\eta = 5.023 T^{0.647}$$

in which the units are in micropoises if T is in degrees Kelvin. The results are in good agreement with the theoretical values. The estimated error is certainly less than 10% even at high f/p values.

2. Sound Absorption Measurements in Helium at 76.7 °K.

The apparatus was next tested for proper operation at liquid nitrogen temperature. Due to the low atmospheric

PROCEEDINGS OF THE

ANNUAL MEETING OF THE AMERICAN ASSOCIATION OF GEOLOGICAL AND NATURAL HISTORIANS

HELD AT THE UNIVERSITY OF CHICAGO, CHICAGO, ILL., SEPTEMBER 1-10, 1875

REPORT OF THE SECRETARY, J. D. COOK, AND OF THE COMMITTEES ON THE PROCEEDINGS OF THE MEETING, AND ON THE PAPERS PRESENTED.

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OFFICE OF THE SECRETARY, 101 N. LAKE STREET, CHICAGO, ILL.

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Table 1
Data necessary for the computation of
 α class and ϵ .

Table 1

Gas	T	h	φ	C ⁱ /R	C _v ^o /R	C _p ^o /R	γ	V _o	f	(α p) _{class}	ϵ
	^o K	μ P	$10^6 \frac{\text{cal}}{(\text{cm}^3 \text{Ksec})}^{-1}$					m/sec	Mc/sec	cm ⁻¹ atm	
He	296	119	—	—	1.50	2.50	1.66	995	3.00	0.495	—
He	76.7	82.8	—	—	1.50	2.50	1.66	515	2.85	0.358	—
n-H ₂	76.7	34.8	119	0.08	1.58	2.58	1.63	720	2.85	0.103	0.02
p-H ₂	76.7	34.8	125	0.29	1.79	2.79	1.55	703	2.85	0.103	0.06

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Table 2
Absorption in Helium at 296°K.

Table 2

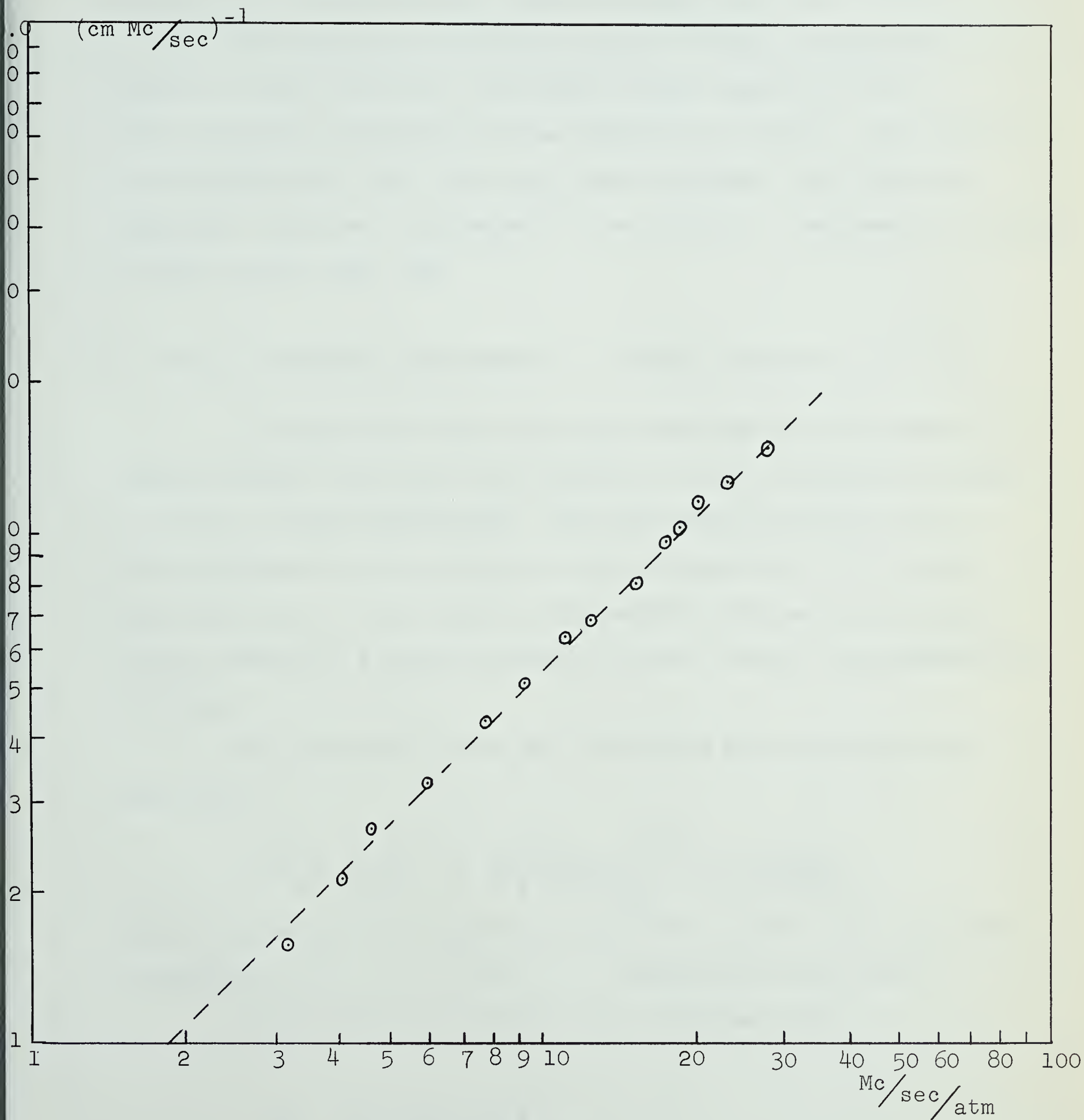
$1/p$ Atm^{-1}	f/p Mc/sec/atm	α cm^{-1}	α/f $(\text{cm Mc/sec})^{-1}$
1.05	3.15	0.48	0.156
1.34	4.02	0.63	0.210
1.53	4.59	0.805	0.264
1.96	5.88	0.97	0.324
2.57	7.71	1.28	0.430
3.08	9.24	1.52	0.510
3.70	11.1	1.90	0.630
4.10	12.3	2.05	0.680
5.05	15.15	2.40	0.800
5.80	17.4	2.90	0.970
6.20	18.6	3.08	1.03
6.70	20.1	3.50	1.17
7.65	22.95	3.80	1.27
9.20	27.6	4.45	1.48

Table

Year	Age	Sex	Weight
1900	1	Male	100
1901	2	Female	110
1902	3	Male	120
1903	4	Female	130
1904	5	Male	140
1905	6	Female	150
1906	7	Male	160
1907	8	Female	170
1908	9	Male	180
1909	10	Female	190
1910	11	Male	200
1911	12	Female	210
1912	13	Male	220
1913	14	Female	230
1914	15	Male	240
1915	16	Female	250
1916	17	Male	260
1917	18	Female	270
1918	19	Male	280
1919	20	Female	290
1920	21	Male	300
1921	22	Female	310
1922	23	Male	320
1923	24	Female	330
1924	25	Male	340
1925	26	Female	350
1926	27	Male	360
1927	28	Female	370
1928	29	Male	380
1929	30	Female	390
1930	31	Male	400
1931	32	Female	410
1932	33	Male	420
1933	34	Female	430
1934	35	Male	440
1935	36	Female	450
1936	37	Male	460
1937	38	Female	470
1938	39	Male	480
1939	40	Female	490
1940	41	Male	500
1941	42	Female	510
1942	43	Male	520
1943	44	Female	530
1944	45	Male	540
1945	46	Female	550
1946	47	Male	560
1947	48	Female	570
1948	49	Male	580
1949	50	Female	590
1950	51	Male	600
1951	52	Female	610
1952	53	Male	620
1953	54	Female	630
1954	55	Male	640
1955	56	Female	650
1956	57	Male	660
1957	58	Female	670
1958	59	Male	680
1959	60	Female	690
1960	61	Male	700
1961	62	Female	710
1962	63	Male	720
1963	64	Female	730
1964	65	Male	740
1965	66	Female	750
1966	67	Male	760
1967	68	Female	770
1968	69	Male	780
1969	70	Female	790
1970	71	Male	800
1971	72	Female	810
1972	73	Male	820
1973	74	Female	830
1974	75	Male	840
1975	76	Female	850
1976	77	Male	860
1977	78	Female	870
1978	79	Male	880
1979	80	Female	890
1980	81	Male	900
1981	82	Female	910
1982	83	Male	920
1983	84	Female	930
1984	85	Male	940
1985	86	Female	950
1986	87	Male	960
1987	88	Female	970
1988	89	Male	980
1989	90	Female	990
1990	91	Male	1000
1991	92	Female	1010
1992	93	Male	1020
1993	94	Female	1030
1994	95	Male	1040
1995	96	Female	1050
1996	97	Male	1060
1997	98	Female	1070
1998	99	Male	1080
1999	100	Female	1090
2000	101	Male	1100
2001	102	Female	1110
2002	103	Male	1120
2003	104	Female	1130
2004	105	Male	1140
2005	106	Female	1150
2006	107	Male	1160
2007	108	Female	1170
2008	109	Male	1180
2009	110	Female	1190
2010	111	Male	1200
2011	112	Female	1210
2012	113	Male	1220
2013	114	Female	1230
2014	115	Male	1240
2015	116	Female	1250
2016	117	Male	1260
2017	118	Female	1270
2018	119	Male	1280
2019	120	Female	1290
2020	121	Male	1300
2021	122	Female	1310
2022	123	Male	1320
2023	124	Female	1330
2024	125	Male	1340
2025	126	Female	1350
2026	127	Male	1360
2027	128	Female	1370
2028	129	Male	1380
2029	130	Female	1390
2030	131	Male	1400
2031	132	Female	1410
2032	133	Male	1420
2033	134	Female	1430
2034	135	Male	1440
2035	136	Female	1450
2036	137	Male	1460
2037	138	Female	1470
2038	139	Male	1480
2039	140	Female	1490
2040	141	Male	1500
2041	142	Female	1510
2042	143	Male	1520
2043	144	Female	1530
2044	145	Male	1540
2045	146	Female	1550
2046	147	Male	1560
2047	148	Female	1570
2048	149	Male	1580
2049	150	Female	1590
2050	151	Male	1600
2051	152	Female	1610
2052	153	Male	1620
2053	154	Female	1630
2054	155	Male	1640
2055	156	Female	1650
2056	157	Male	1660
2057	158	Female	1670
2058	159	Male	1680
2059	160	Female	1690
2060	161	Male	1700
2061	162	Female	1710
2062	163	Male	1720
2063	164	Female	1730
2064	165	Male	1740
2065	166	Female	1750
2066	167	Male	1760
2067	168	Female	1770
2068	169	Male	1780
2069	170	Female	1790
2070	171	Male	1800
2071	172	Female	1810
2072	173	Male	1820
2073	174	Female	1830
2074	175	Male	1840
2075	176	Female	1850
2076	177	Male	1860
2077	178	Female	1870
2078	179	Male	1880
2079	180	Female	1890
2080	181	Male	1900
2081	182	Female	1910
2082	183	Male	1920
2083	184	Female	1930
2084	185	Male	1940
2085	186	Female	1950
2086	187	Male	1960
2087	188	Female	1970
2088	189	Male	1980
2089	190	Female	1990
2090	191	Male	2000
2091	192	Female	2010
2092	193	Male	2020
2093	194	Female	2030
2094	195	Male	2040
2095	196	Female	2050
2096	197	Male	2060
2097	198	Female	2070
2098	199	Male	2080
2099	200	Female	2090
2100	201	Male	2100

Figure 9
 α/f vs f/p for helium at 296 °K
○ Experimental points
- - - - Theoretical curve

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pressure in this laboratory, liquid nitrogen boils at 76.7° K.

The results in helium at liquid nitrogen temperature appear in fig. 10 and the individual values figure in table 3. Again, they are compared with the theoretical values. The value of the viscosity was taken this time from Corremans, van Itterbeek, Beenakker, Knaap and Zandbergen¹². See table 1. The general accuracy is again better than 10%.

3. Sound Absorption Measurements in Normal Hydrogen at 76.7° K.

Although both para and ortho modifications are present in normal hydrogen, the gas should exhibit a single relaxation process at liquid nitrogen temperature, since the first excited level of ortho hydrogen is not populated at that temperature. As can be seen from fig. 11, and table 4, the results obtained in this laboratory indeed fit a single relaxation curve, within the experimental accuracy.

The theoretical curve was calculated from the following expression

$$\alpha'/f = \frac{1}{V_0} \left(1 - \epsilon \frac{\omega^2 \tau_{ps}^2}{1 + \omega^2 \tau_{ps}^2} \right)^{-1/2} \pi \epsilon \frac{\omega \tau_{ps}}{1 + \omega^2 \tau_{ps}^2}$$

which is obtained from equations (1-80) and (1-81). This curve has a maximum at $\tau_{ps} = (1 - \epsilon)^{-1/4}$. (Herzfeld and Litovitz)¹³

The relaxation strength was calculated from

$$\epsilon = \frac{C' (C_p^0 - C_v^0)}{C_v^0 (C_p^0 - C')}$$

1. The first part of the paper is devoted to a general discussion of the problem.

2. In the second part, we consider the case of a single particle.

3. The third part is devoted to the case of a system of particles.

4. In the fourth part, we consider the case of a continuous medium.

5. The fifth part is devoted to the case of a system of continuous media.

6. In the sixth part, we consider the case of a system of continuous media.

7. The seventh part is devoted to the case of a system of continuous media.

8. In the eighth part, we consider the case of a system of continuous media.

9. The ninth part is devoted to the case of a system of continuous media.

10. In the tenth part, we consider the case of a system of continuous media.

11. The eleventh part is devoted to the case of a system of continuous media.

12. In the twelfth part, we consider the case of a system of continuous media.

13. The thirteenth part is devoted to the case of a system of continuous media.

14. In the fourteenth part, we consider the case of a system of continuous media.

15. The fifteenth part is devoted to the case of a system of continuous media.

16. In the sixteenth part, we consider the case of a system of continuous media.

17. The seventeenth part is devoted to the case of a system of continuous media.

18. In the eighteenth part, we consider the case of a system of continuous media.

19. The nineteenth part is devoted to the case of a system of continuous media.

20. In the twentieth part, we consider the case of a system of continuous media.

21. The twenty-first part is devoted to the case of a system of continuous media.

22. In the twenty-second part, we consider the case of a system of continuous media.

Table 3

Absorption in helium at 76.7°K .



Table 3

$1/p$ Atm^{-1}	f/p $\text{Mc}/\text{sec}/\text{atm}$	α cm^{-1}	α/f $(\text{cm Mc}_{\text{sec}})^{-1}$
1.48	4.23	0.52	0.182
2.47	7.02	0.885	0.310
3.30	9.40	1.14	0.400
4.48	12.8	1.80	0.630
5.43	15.4	2.08	0.730
5.80	16.5	2.22	0.778
6.28	17.9	2.52	0.884
6.84	19.6	2.64	0.925
7.23	20.7	2.76	0.968
8.05	22.9	3.07	1.07
8.58	24.4	3.20	1.12
9.30	26.5	3.54	1.24
10.7	30.5	4.13	1.45
12.4	35.4	4.22	1.48
14.8	42.2	5.15	1.81

Figure 10
 α/f vs f/p for helium at 76.7° K
⊙ Experimental points
- - - - Theoretical curve

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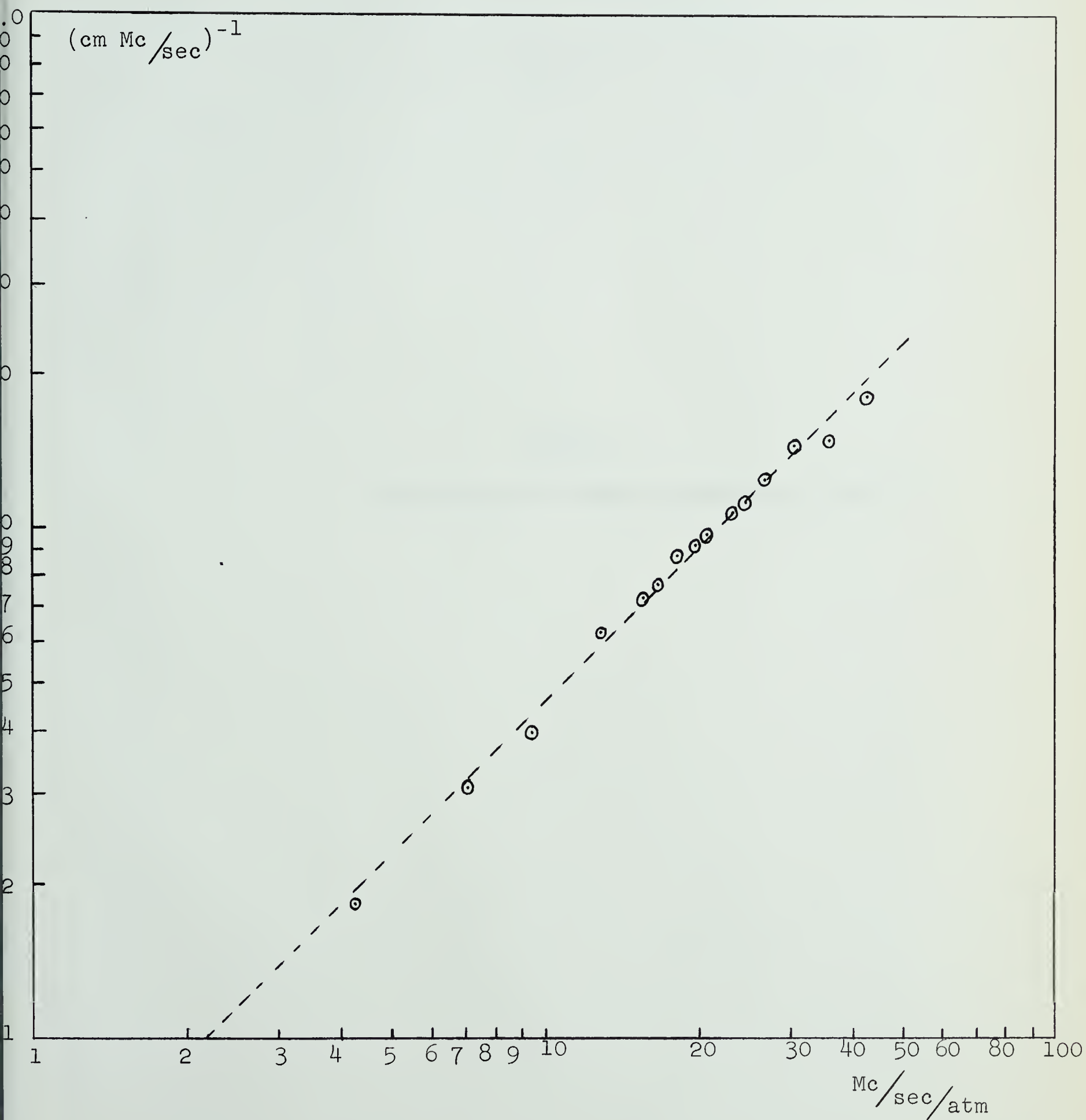




Table 4

Absorption in normal hydrogen at 76.7° K

Table 4

f/p Mc/sec/atm	α_{class} cm^{-1}	α_{exp} cm^{-1}	α' cm^{-1}	α'/f $(\text{cm Mc/sec})^{-1}$
2.42	0.086	0.755	0.67	0.234
3.30	0.118	1.08	0.96	0.330
3.80	0.137	1.09	0.95	0.333
5.02	0.185	1.40	1.22	0.428
5.48	0.194	1.34	1.15	0.403
5.90	0.213	1.46	1.25	0.439
6.62	0.234	1.46	1.23	0.430
7.12	0.258	1.56	1.28	0.450
7.65	0.276	1.61	1.33	0.465
8.15	0.294	1.50	1.21	0.425
8.73	0.316	1.51	1.20	0.421
8.80	0.318	1.60	1.28	0.450
9.60	0.346	1.62	1.27	0.445
10.3	0.372	1.58	1.21	0.423
11.5	0.418	1.63	1.21	0.423
13.0	0.463	1.52	1.06	0.372
16.0	0.570	1.48	0.91	0.318
20.3	0.730	1.54	0.81	0.284
32.3	1.14	1.81	0.67	0.235
45.2	1.60	2.06	0.46	0.160

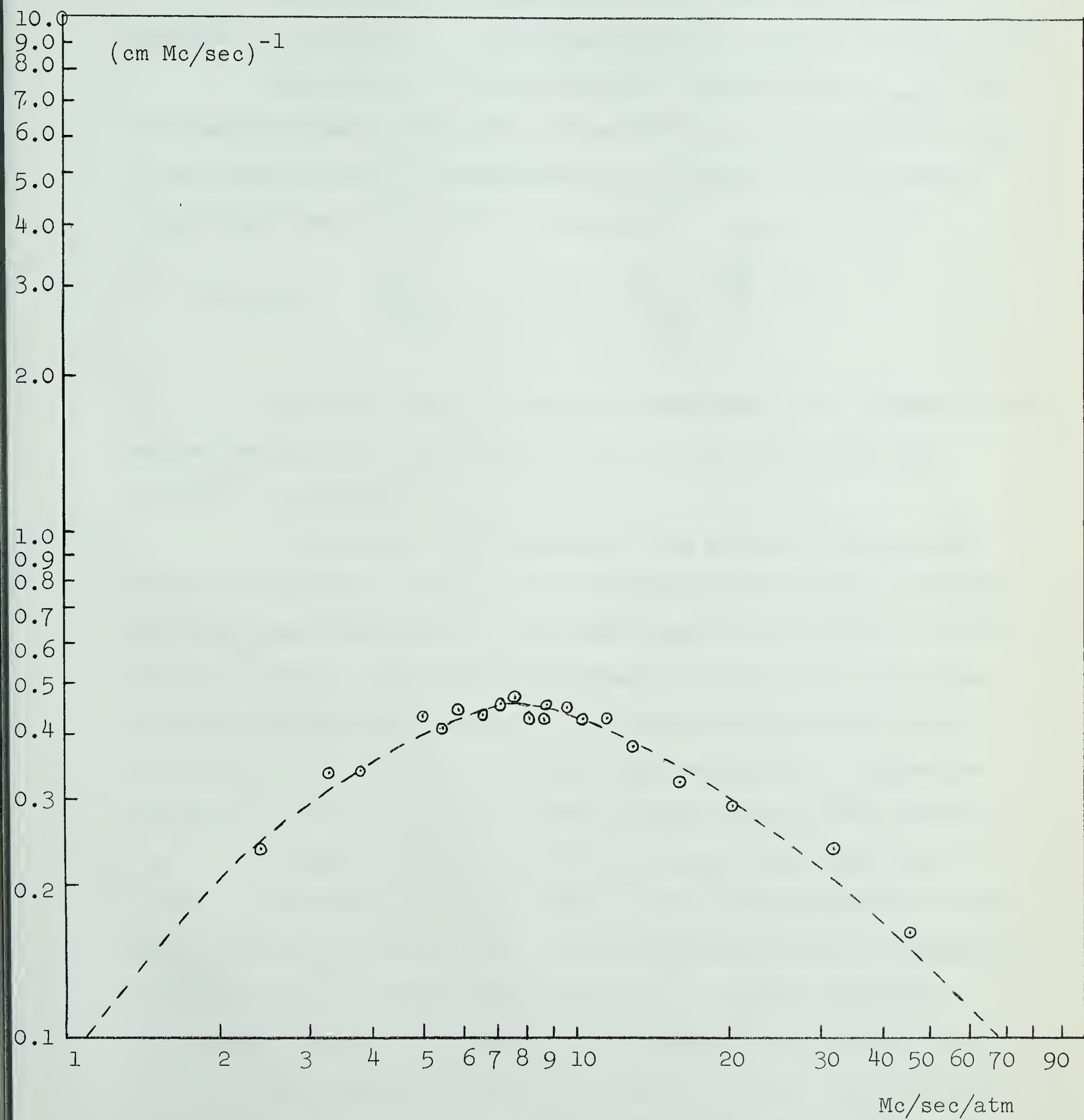
Figure 11

α'/f vs f/p for normal hydrogen at 76.7° K

○ Experimental points

- - - - Theoretical curve calculated for $\epsilon = 0.02$

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The values of the specific heats were taken from Farkas¹⁴. See table 1. For normal hydrogen at 76.7° K, $\epsilon = 0.02$.

The value of α' was obtained by subtracting α_{class} from the experimentally determined value of α which is denoted by α_{exp} . Since hydrogen is not a monatomic gas, equation (1-59) rather than (1-58) had to be used in calculating α_{class} :

$$\alpha_{\text{class}} = \frac{2\pi^2}{\gamma p_0 v_0} \left[\frac{4}{3} \eta + \frac{(\gamma - 1)}{c_p^0} \right] f^2$$

The data for the viscosity were taken from Coremans et al., while the data for the thermal conductivity were taken from Ubbink¹⁵. See table 1.

In general, the accuracy of the results seems poorer than in the case of helium. It is reasonable however, to assume that α_{exp} was determined to the same degree of accuracy as in the case of helium. The higher discrepancy between the theoretical and the experimental values of α'/f is due to the fact that α' represents only a fraction of the total absorption. Therefore, an error of 10% on α_{exp} constitutes a higher percentage error on α' . For example, at 20.3 Mc/sec-atm, α_{exp} was found equal to 1.54. A 10% error is 0.15. Since α' is obtained by subtracting α_{class} from α_{exp} , α' is 0.81. The absolute error 0.15 represents a relative error of more than 18% on α' . As the classical absorption becomes more important, α' represents a smaller fraction of the total absorption and the accuracy to which α' can be determined becomes lesser. Taking this fact into account, the results

in fig.11 agree with the theoretical curve well within the experimental error.

The maximum of the theoretical curve which best fits the experimental points is located at 7.65 Mc/sec-atm. At this point, $\omega \hat{\tau}_{pS} = (1 - 0.02)^{-1/4}$. Since $\hat{\tau}_{pS}$ is the adiabatic relaxation time at unit pressure, the maximum of the relaxational absorption occurs at a frequency

$$2\pi \times 7.65 \times 10^6 \text{ sec}^{-1}.$$

From this, $\hat{\tau}_{pS}$ is found to be equal to

$$\frac{(1 - 0.02)^{-1/4}}{2\pi \times 7.65 \times 10^6} = 2.11 \times 10^{-8} \text{ sec.}$$

The isothermal or true relaxation time can be calculated from the following relation

$$\frac{\hat{\tau}_{pS}}{\hat{\tau}_{pT}} = \frac{C_p^\infty}{C_p^0}.$$

$$\text{With } C_p^0 = 2.58R \quad \text{and } C_p^\infty = 2.50R, \quad \hat{\tau}_{pT} = 2.17 \times 10^{-8} \text{ sec.}$$

4. Sound Absorption Measurements in Para-enriched Hydrogen.

An attempt was made at producing pure para hydrogen. Normal hydrogen was liquefied in the presence of a catalyst (hydrous ferric oxide). Since liquid hydrogen is not available in this

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laboratory, conversion was effected at a temperature in the neighbourhood of 20° K, above the surface of the liquid inside a Dewar containing liquid helium. A few reasons could have contributed to the lack of success of this experiment; the catalyst for instance was probably not activated sufficiently, and the liquified hydrogen may not have been in contact with the catalyst for a long enough time.

Nevertheless, a definite enrichment in para content was achieved, and sound absorption measurements were taken in this para-ortho mixture. The results are shown in fig. 4 and the individual values figure in table 5.

From the maximum value of α'/f , the relaxation strength was found to be equal to 0.045. The theoretical values of the relaxation strength of normal and pure para hydrogen are 0.02 and 0.06 respectively. A relaxation strength of 0.045 corresponds to a rotational specific heat C' of 0.19 R.

The maximum of the absorption curve occurs at a f/p value of 8.1 Mc/sec-atm. From this, τ_{pS} is found to be equal to 1.96×10^{-8} sec. The isothermal relaxation time in this case is

$$\tau_{pT} = 1.96 \times \frac{2.69}{2.50} \times 10^{-8} \text{ sec}$$

or
$$\tau_{pT} = 2.11 \times 10^{-8} \text{ sec,}$$

which agrees within a few percent with the isothermal relaxation time for the $0 \longrightarrow 2$ transition found in normal hydrogen.

It is of interest to compare the present results with those obtained by other authors. Sound absorption measurements

Table 5

Absorption in para-enriched hydrogen at 76.7° K

Table 5

f/p $\text{Mc}_{\text{sec atm}}$	α_{class} cm^{-1}	α_{exp} cm^{-1}	α' cm^{-1}	α'/f $(\text{cm Mc}_{\text{sec}})^{-1}$
3.03	0.109	1.72	1.61	0.57
4.28	0.154	2.40	2.25	0.79
5.90	0.214	2.68	2.47	0.87
7.10	0.258	2.93	2.67	0.94
8.10	0.293	3.18	2.89	1.01
9.60	0.345	3.08	2.74	0.96
10.6	0.375	3.08	2.71	0.95
11.5	0.415	3.10	2.69	0.92
12.9	0.465	2.83	2.37	0.83
14.6	0.530	2.98	2.45	0.86
16.9	0.610	2.84	2.23	0.78
24.6	0.892	2.67	1.78	0.62
30.9	1.11	2.33	1.22	0.43

Table 1

Year	1980	1981	1982	1983	1984
1980	100	100	100	100	100
1981	100	100	100	100	100
1982	100	100	100	100	100
1983	100	100	100	100	100
1984	100	100	100	100	100
1985	100	100	100	100	100
1986	100	100	100	100	100
1987	100	100	100	100	100
1988	100	100	100	100	100
1989	100	100	100	100	100
1990	100	100	100	100	100
1991	100	100	100	100	100
1992	100	100	100	100	100
1993	100	100	100	100	100
1994	100	100	100	100	100
1995	100	100	100	100	100
1996	100	100	100	100	100
1997	100	100	100	100	100
1998	100	100	100	100	100
1999	100	100	100	100	100
2000	100	100	100	100	100
2001	100	100	100	100	100
2002	100	100	100	100	100
2003	100	100	100	100	100
2004	100	100	100	100	100
2005	100	100	100	100	100
2006	100	100	100	100	100
2007	100	100	100	100	100
2008	100	100	100	100	100
2009	100	100	100	100	100
2010	100	100	100	100	100
2011	100	100	100	100	100
2012	100	100	100	100	100
2013	100	100	100	100	100
2014	100	100	100	100	100
2015	100	100	100	100	100
2016	100	100	100	100	100
2017	100	100	100	100	100
2018	100	100	100	100	100
2019	100	100	100	100	100
2020	100	100	100	100	100

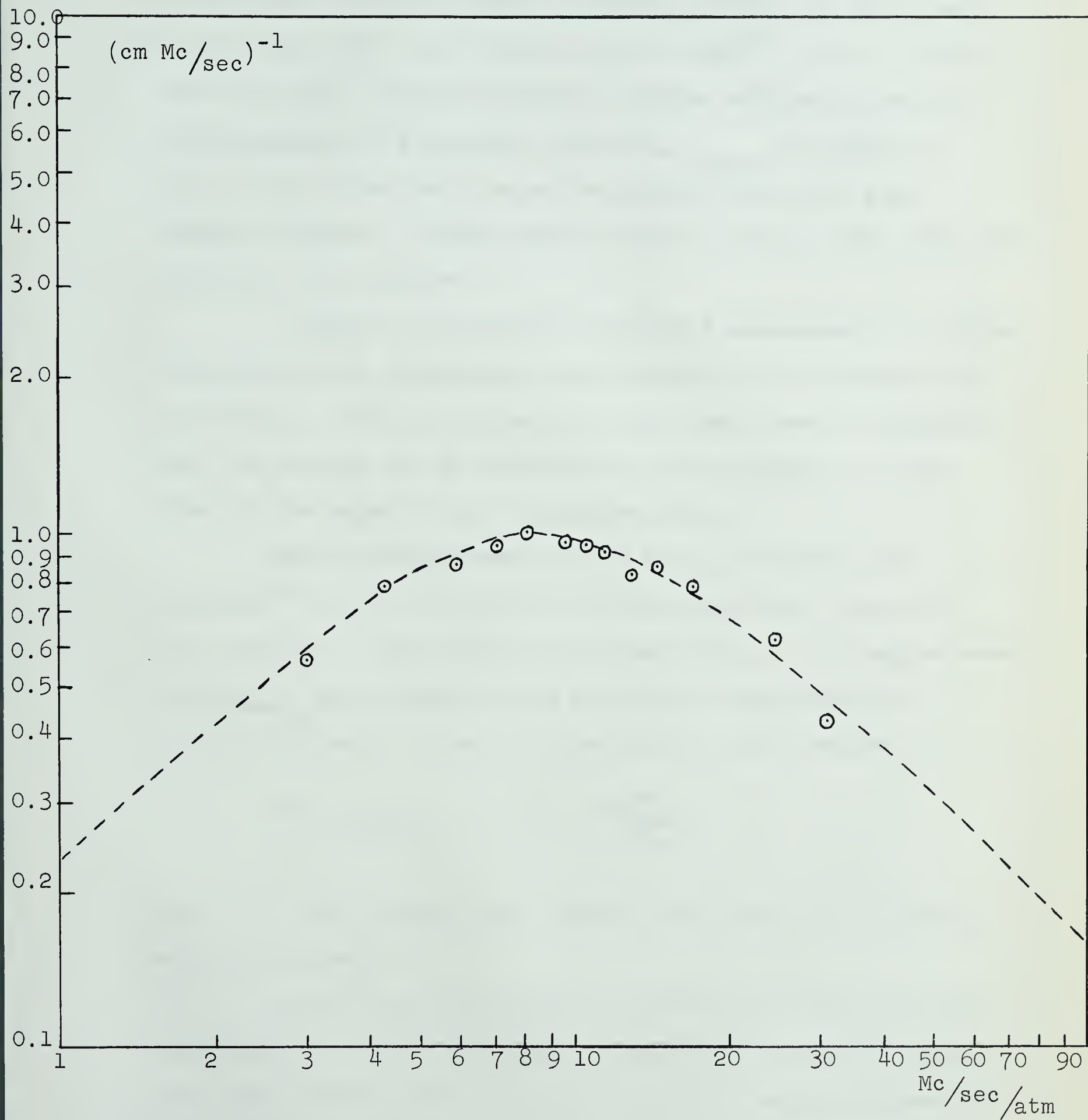
Figure 12

d'/f vs f/p for para-enriched hydrogen at 76.7° K

⊙ Experimental points

- - - - Theoretical curve calculated for $\epsilon = 0.045$

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in hydrogen have been taken by several authors for many years. In 1937 and 1938, van Itterbeek and Mariens¹⁶, and van Itterbeek and Thys¹⁷ reported results obtained with an ultrasonic interferometer in the region where $\alpha/\alpha_{\text{class}}$ is constant, i.e. at f/p values well below the region where α'/f goes through a maximum. These results however did not show the right variation with pressure.

Stewart and Stewart¹⁸ conducted measurements in normal hydrogen with an ultrasonic interferometer at a frequency of 1.95 Mc/sec. With an accuracy of 20%, their results indicated that the maximum of the absorption curve occurred at a f/p value of the order of 10 -- 12 Mc/sec/atm.

Improved measurements taken by van Itterbeek and Verhaegen¹⁹ at 79.5 °K and 90 °K showed the right variation with pressure. These results obtained again in the region where $\alpha/\alpha_{\text{class}}$ is constant, yield a value of approximately 2.5×10^{-8} sec. It can be shown that in that region,

$$\alpha/\alpha_{\text{class}} = 1 + 0.067 \frac{\tau_{pT}}{\tau_c}$$

where τ_c is the average time between collisions at a pressure of one atmosphere.

Using the pulse method, Parbrook and Tempest²⁰ took measurements in normal hydrogen at 298 °K. The results show a lower and broader curve which does not fit a single-relaxation curve. This is to be expected since at room temperature, normal hydrogen has five excited levels giving rise to four

possible transitions, with possibly four different relaxation times.

All measurements made in either pure para hydrogen or in normal hydrogen above 150 °K have to be interpreted in terms of two or more relaxation processes. This necessitates the use of many assumptions. In order to analyse the results which he obtained at 293 °K in normal hydrogen, Sessler²¹ made the following assumptions: the relaxation time for the $1 \longrightarrow 3$ transition is 1.5 times that for the $0 \longrightarrow 2$ transition, and the relaxation time for the $3 \longrightarrow 5$ transition is the same as that for the $1 \longrightarrow 3$ transition. He assumed moreover that all transitions occur independently. From this, Sessler found that at room temperature the isothermal relaxation time for the $0 \longrightarrow 2$ transition (τ_{02}) is 1.29×10^{-8} sec.

Geide²² who took measurements in pure para hydrogen at 293 °K, also assumed that the $2 \longrightarrow 4$ transition occurs independently of the $0 \longrightarrow 2$ transition. With this assumption, his measurements yield a value of $\tau_{02} = 1.21 \times 10^{-8}$ sec.

It must be emphasized that these relaxation times (1.29 and 1.21×10^{-8} sec) were obtained from measurements taken at room temperature, and it is reasonable to expect that they should differ from the relaxation times obtained from measurements taken at lower temperatures.

The following considerations will give an idea of the way in which the relaxation times can be expected to vary with temperature.

As was pointed out at the end of the first chapter, the isothermal relaxation time at unit concentration is the reciprocal of the sum of the reaction rates. In para hydrogen

$$\tau_T^{-1} = R_{02} + R_{20} .$$

The relation between R_{02} and R_{20} is provided by the condition of statistical equilibrium,

$$R_{20} = R_{02} \left(\frac{g_0}{g_2} \right) e^{E/kT}$$

where g_0 and g_2 are the statistical weights of the rotational levels $J=0$ and $J=2$ respectively. E is the energy difference between the energy of the state $J=2$ and $J=0$.

Now, the reaction rate is proportional to the number of molecules striking unit area per unit time at pressure p and temperature T . From kinetic theory, this number of molecules is $\left(\frac{8}{\pi m k T} \right)^{1/2} p$. The reaction rate is also proportional to the efficiency of the collisions in inducing rotational energy transfer. The efficiency of the collisions is expressed in terms of an averaged inelastic cross section for the $0 \longrightarrow 2$ transition at temperature T and pressure p . This cross section is denoted by $\langle Q_{02} \rangle_T$. Therefore,

$$R_{02} = \left(\frac{8}{\pi m k T} \right)^{1/2} p \langle Q_{02} \rangle_T ,$$

and

$$\tau_T^{-1} = \left(\frac{8}{\pi m k T} \right)^{1/2} p \langle Q_{02} \rangle_T \left(1 + \frac{g_0}{g_2} e^{-E/kT} \right).$$

$\langle Q_{02} \rangle_T$ is obtained by averaging $Q(K)$, the inelastic cross section corresponding to a wave number K , over the Maxwellian distribution of wave numbers K at temperature T .

Various authors^{25, 26, 27} have calculated $Q(K)$ for different forms of the intermolecular potential. Using a Morse-type potential with two angle-dependent terms, Davison²⁵ has computed the values of $Q(K)$ as function of K^2 . His results may be approximated as follows:

$$Q(K) = c (K^2 - K_0^2)$$

where $c = 0.043$ and $K_0^2 = 6$ in atomic units. In this system of units, Planck's constant is equal to 2π .

Averaged over the Maxwellian distribution of wave numbers at temperature T , $Q(K)$ yields

$$\langle Q_{02} \rangle_T = \exp(-E/kT) 4cmkT \left(1 + E/2kT \right).$$

Inserting $\langle Q_{02} \rangle_T$ into the expression for τ_T^{-1} , one gets

$$\tau_T^{-1} = \left(\frac{8}{\pi m k T} \right)^{1/2} p \left[\frac{g_0}{g_2} + \exp(-E/kT) \right] 4cmkT \left(1 + E/2kT \right).$$

Above 150 °K, the factor $T^{-1/2}$ is dominant and one should expect the relaxation times to decrease as the temperature

$$f(x) = \frac{1}{2} \ln(x^2 + 1) - \frac{1}{2} \ln(x^2 - 1) = \frac{1}{2} \ln \left(\frac{x^2 + 1}{x^2 - 1} \right)$$

The function $f(x)$ is defined for $x > 1$ and $x < -1$.
 The derivative of $f(x)$ is $f'(x) = \frac{1}{x^2 - 1}$.
 The function $f(x)$ is increasing for $x > 1$ and decreasing for $x < -1$.
 The function $f(x)$ has a vertical asymptote at $x = 1$ and $x = -1$.
 The function $f(x)$ has a horizontal asymptote at $y = 0$.

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 The function $f(x)$ has a vertical asymptote at $x = 1$ and $x = -1$.
 The function $f(x)$ has a horizontal asymptote at $y = 0$.

increases.

A rough calculation shows that the relaxation time at room temperature should be approximately half the relaxation time at liquid nitrogen temperature.

Indeed,

at 80 °K, $E/kT \approx 1$ and $e^{-1} \approx 0.37$;

at 300 °K, $E/kT \approx 0.3$ and $e^{-0.3} \approx 0.72$;

$g_0 = 1$ and $g_2 = 3$.

Therefore,

$$\frac{\tau_{300}}{\tau_{80}} \approx \left(\frac{80}{300} \right)^{1/2} \times \frac{(0.33 + 0.37)}{(0.33 + 0.72)} \times \frac{(1 + 0.5)}{(1 + 0.15)}$$

$$\frac{\tau_{300}}{\tau_{80}} \approx 1/2 \quad .$$

The relaxation time deduced from Sessler's measurements at room temperature (1.2×10^{-8} sec) is roughly half the relaxation time deduced from measurements at liquid nitrogen temperature (2.1×10^{-8} sec).

The computation of $\langle Q_{O_2} \rangle_T$ involves many assumptions and especially, the use of certain adjustable parameters. One may question the validity of the assumptions made, as well as the values of the parameters used. The best way of verifying the reliability of the theory consists of comparing the theoretical and the experimental results over a wide range of temperature. In that respect, $\hat{\tau}_T$ is not the best quantity to consider for making comparisons, because the temperature dependence of

τ_T is affected by the fact that τ_T depends on the number of molecules hitting unit area per unit time. As seen previously, the relaxation time is inversely proportional to the number of molecules hitting unit area per unit time; it is thus proportional to the mean time between collisions τ_c . At pressure p ,

$$\tau_c^{-1} = \pi \sigma^2 \left(\frac{8}{\pi m k T} \right)^{1/2} p$$

where σ is the diameter of the molecules treated as hard spheres.

Consequently, the collision number $Z = \tau_T / \tau_c$ is better suited for comparisons between theory and experiment. Using the expressions for τ_T^{-1} and τ_c^{-1} ,

$$Z^{-1} = \frac{\langle Q_{02} \rangle_T}{\pi \sigma^2} \left(1 + \frac{g_0}{g_2} e^{E/kT} \right).$$

Since $\langle Q_{02} \rangle_T$ results from averaging $Q(K)$ over the Maxwellian distribution of the wave numbers at temperature T , it can be written as

$$\langle Q_{02} \rangle_T = \exp(-E/kT) I(T)$$

where $I(T)$ stands for an integral which depends on the form of the intermolecular potential. With $\langle Q_{02} \rangle_T$ expressed as above,

$$Z^{-1} = \frac{1}{\pi \sigma^2} \left[\frac{g_0}{g_2} + \exp(-E/kT) \right] I(T).$$

Therefore, the temperature dependence of Z is seen to be mostly determined by the temperature dependence of $I(T)$.

1. The first part of the paper is devoted to the study of the

properties of the function $f(x)$ defined by the equation

$$f(x) = \int_0^x \frac{1}{1+t^2} dt$$

and to the study of the function $F(x)$ defined by the equation

$$F(x) = \int_0^x \frac{1}{1+t^2} dt$$

and to the study of the function $G(x)$ defined by the equation

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and to the study of the function $H(x)$ defined by the equation

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$$N(x) = \int_0^x \frac{1}{1+t^2} dt$$

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$$P(x) = \int_0^x \frac{1}{1+t^2} dt$$

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$$Q(x) = \int_0^x \frac{1}{1+t^2} dt$$

and to the study of the function $R(x)$ defined by the equation

$$R(x) = \int_0^x \frac{1}{1+t^2} dt$$

Taking $I(T) = 4cmkT (1 + E/2kT)$ as before,

$$\frac{Z_{300}^{-1}}{Z_{80}^{-1}} \approx \frac{(0.33 + 0.72)}{(0.33 + 0.37)} \times \frac{(1 + 0.15)}{(1 + 0.5)} \times \frac{300}{80}$$

$$\frac{Z_{80}}{Z_{300}} \approx 4.$$

This value is in good agreement with the experimental results:

$$\text{at } 300^\circ\text{K}, \tau_c = 0.7 \times 10^{-10} \text{ sec,}$$

$$\text{at } 80^\circ\text{K}, \tau_c = 0.3 \times 10^{-10} \text{ sec.}$$

Consequently, using Sessler's value of the relaxation time at room temperature,

$$Z_{300} \approx \frac{1.2 \times 10^{-8}}{0.7 \times 10^{-10}} \approx 170 \text{ collisions.}$$

At liquid nitrogen temperature,

$$Z_{80} \approx \frac{2.1 \times 10^{-8}}{0.3 \times 10^{-10}} \approx 700 \text{ collisions.}$$

These values of the collision numbers are in good agreement within an order of magnitude, with the theoretical values deduced from the calculations of Sluijter et al.²⁴, and those of Brout³⁰.

The only results obtained in hydrogen below 150 °K, which are reported in the literature, are those of Sluijter, Knaap and Beenakker^{8, 23, 24}. From their measurements taken in

pure para hydrogen at 77.3 °K and 90 °K, the values of τ_{02} are found to be 2.2×10^{-8} sec and 2.1×10^{-8} sec respectively.

Measurements by Sluijter et al. in normal hydrogen at 77.3 °K, however indicated that the maximum of the relaxational absorption occurred at approximately 11 Mc/sec/atm or even higher. This indicated a value of $\tau_{02} = 1.4 \times 10^{-8}$ sec.

This important difference between the relaxation time of para hydrogen at 100% concentration, and the relaxation time at 25% concentration (2.2×10^{-8} sec and 1.4×10^{-8} sec respectively), raises the question as to whether τ_{02} is the same when collisions occur between para molecules only, as it is when para and ortho molecules collide together. A shorter relaxation time in the case of normal hydrogen would mean that ortho molecules are more efficient than para molecules at inducing rotational energy transfer during a collision.

The present results seem to indicate that such is not the case, since the values of τ_{02} obtained from measurements in normal hydrogen are in good agreement with the value of τ_{02} obtained by Sluijter et al. from measurements in pure para hydrogen.

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CONCLUSION

By means of the pulse technique at liquid nitrogen temperature, the sound absorption was measured in normal and para-enriched hydrogen at frequency-to-pressure ratios in the range 3 to 50 Mc/sec/atm.

These measurements are in good agreement with the theory first proposed by Herzfeld and Rice, both as to their dependence on f/p and their absolute magnitude.

The peak in the absorption curve, referred to a pressure of one atmosphere, occurs when $\omega^{-1} = \tau_{pS}$, the adiabatic relaxation time. From this, $\tau_T = \tau_{O_2}$ was deduced, and the value obtained was $\tau_{O_2} = 2.1 \times 10^{-8}$ sec, for both para-enriched and normal hydrogen.

This value of τ_{O_2} is in good agreement with that deduced from measurements taken by the direct method at the same temperature, in pure para hydrogen. It differs however from the value of τ_{O_2} deduced from measurements taken by other authors in normal hydrogen at the same temperature.

Although no claim is made at a particularly high experimental accuracy, the fact that the number of measurements (i.e. the number of points on the absorption curve) taken in normal hydrogen at liquid nitrogen temperature and reported in this thesis, is considerably larger than the number of similar measurements taken and reported by any other authors, may confer to the present results a certain degree of reliability.

CHAPTER I

The first part of the book is devoted to the study of the properties of the function $f(x)$ and its derivatives. In the second part, we shall consider the problem of the determination of the function $f(x)$ from its values at a finite number of points.

§ 1. The function $f(x)$ and its derivatives.

Let $f(x)$ be a function defined on the interval (a, b) . We say that $f(x)$ is differentiable at the point x if the limit $\lim_{h \rightarrow 0} \frac{f(x+h) - f(x)}{h}$ exists. This limit is called the derivative of $f(x)$ at the point x and is denoted by $f'(x)$.

The derivative of $f(x)$ at the point x is denoted by $f'(x)$. The derivative of $f(x)$ at the point x is denoted by $f'(x)$. The derivative of $f(x)$ at the point x is denoted by $f'(x)$.

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The relaxation theory of Herzfeld and Rice is a thermodynamical one and as such can only provide a macroscopic description of the phenomenon involved. A comparison of the relaxation times obtained at liquid nitrogen temperature, with those obtained by other authors at room temperature shows that the microscopic theory predicts quite satisfactorily the temperature variation of the relaxation times. A full verification of the microscopic theory must however await experimental results at other temperatures. Moreover, theories are not precise enough at the present time to give absolute magnitude of the relaxation times to better than within an order of magnitude.

As yet, no reliable information has been obtained about the relaxation time of the $1 \longrightarrow 3$ transition in ortho hydrogen. If measurements could be performed in pure ortho hydrogen, the results would be most interesting. Unfortunately, the production of ortho hydrogen although feasible²⁹, remains difficult especially in view of the large amount of gas necessary to fill a cryostat. Measurements in normal hydrogen or in various mixtures of ortho and para hydrogen between 140 °K and 180 °K could also provide some information about the relaxation time of the ortho modification. Results in ortho hydrogen would constitute another way of verifying the microscopic theories.

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Introduction

The purpose of this study is to investigate the effects of various factors on the growth and development of the human body. The study is divided into two main sections: a theoretical section and a practical section. The theoretical section discusses the various factors that influence growth and development, including genetics, nutrition, and environment. The practical section describes the methods used to collect and analyze data on growth and development. The results of the study are presented in the following sections.

The first section of the study is a review of the literature on growth and development. This section discusses the various factors that influence growth and development, including genetics, nutrition, and environment. The second section of the study is a description of the methods used to collect and analyze data on growth and development. The third section of the study is a presentation of the results of the study. The fourth section of the study is a discussion of the implications of the results for future research and practice.

The study was conducted over a period of six months. Data was collected from a sample of 100 children aged 5 to 12 years. The data was analyzed using statistical methods. The results of the study are presented in the following sections.

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